

1 **DFe patterns impacted by shallow hydrothermal sources along a transect through**  
2 **the Tonga-Kermadec arc**

3 **C. Tilliette<sup>1\*</sup>, V. Taillandier<sup>1</sup>, P. Bouruet-Aubertot<sup>2</sup>, N. Grima<sup>3</sup>, C. Maes<sup>3</sup>, M. Montanes<sup>1</sup>, G.**  
4 **Sarthou<sup>4</sup>, M-E. Vorrath<sup>5</sup>, V. Arnone<sup>6</sup>, M. Bressac<sup>1</sup>, D. González-Santana<sup>4</sup>, F. Gazeau<sup>1</sup> and**  
5 **C. Guieu<sup>1\*</sup>**

6 <sup>1</sup>Sorbonne Université, CNRS, Laboratoire d'Océanographie de Villefranche, LOV, 06230,  
7 Villefranche-sur-Mer, France

8 <sup>2</sup>Sorbonne Université, CNRS, IRD, Laboratoire d'Océanographie et de Climatologie par  
9 Expérimentation et Approche Numérique (LOCEAN), Paris, France

10 <sup>3</sup>Univ Brest, CNRS, IRD, Ifremer, Laboratoire d'Océanographie Physique et Spatiale (LOPS,  
11 UMR 6523), IUEM, Brest, France

12 <sup>4</sup>Univ Brest, CNRS, IRD, Ifremer, LEMAR, F-29280 Plouzane, France

13 <sup>5</sup>Alfred Wegener Institute, Helmholtz Centre for Polar and Marine Research, 27568  
14 Bremerhaven, Germany

15 <sup>6</sup>Instituto de Oceanografía y Cambio Global, IOCAG, Universidad de Las Palmas de Gran  
16 Canaria, ULPGC, 35017 Las Palmas de Gran Canaria, Spain

17 \*Corresponding authors: Chloé Tilliette ([chloe.tilliette@imev-mer.fr](mailto:chloe.tilliette@imev-mer.fr)) and Cécile Guieu  
18 ([cecile.guieu@imev-mer.fr](mailto:cecile.guieu@imev-mer.fr))

19 **Key Points:**

- 20 • Shallow hydrothermal plumes of the Tonga-Kermadec arc are not transported over long  
21 distances, as previously reported for deep plumes.
- 22 • Surface scavenging and photoreduction of stabilizing-complexes mediate the low spatial  
23 dispersion of shallow hydrothermal dissolved iron.
- 24 • Nevertheless, the cumulative impact of multiple sources along the Tonga-Kermadec arc  
25 fertilizes the entire Lau Basin with dissolved iron.

## 26 **Abstract**

27 In the Western Tropical South Pacific, a hotspot of N<sub>2</sub>-fixing organisms has recently been  
28 identified. The survival of these species depends on the availability of dissolved iron (dFe). dFe  
29 was measured along a transect from 175 °E to 166 °W near 19-21 °S. The distribution of dFe  
30 showed high spatial variability: low concentrations (~0.2 nmol kg<sup>-1</sup>) in the South Pacific gyre  
31 and high concentrations (up to 50 nmol kg<sup>-1</sup>) west of the Tonga arc, indicating that this arc is a  
32 clear boundary between iron-poor and iron-rich waters. An optimal multiparameter analysis was  
33 used to distinguish the relative importance of physical transport relative to non-conservative  
34 processes on the observed dFe distribution. This analysis demonstrated that distant sources of  
35 iron play a minor role in its distribution along the transect. The high concentrations observed  
36 were therefore attributed to shallow hydrothermal sources massively present along the Tonga-  
37 Kermadec arc. Nevertheless, in contrast to what has been observed for deep hydrothermal  
38 plumes, our results highlighted the rapid decrease in dFe concentrations near shallow  
39 hydrothermal sources. This is likely due to a shorter residence time of surface water masses  
40 combined with several biogeochemical processes at play (e.g., precipitation, photoreduction,  
41 scavenging, biological uptake). This study clearly highlights the role of shallow hydrothermal  
42 sources on the dFe cycle within the Tonga-Kermadec arc where a strong link to biological  
43 activity in surface waters can be assessed. It also emphasizes the need to consider the impact of  
44 these shallow hydrothermal sources for a better understanding of the global iron cycle.

## 45 **1. Introduction**

46 Iron (Fe) is the fourth most abundant element in the Earth's crust (about 6.7%; Rudnick & Gao,  
47 2003) but it is present at sub-nanomolar concentrations in seawater (< 10<sup>-9</sup> mol L<sup>-1</sup>; Johnson et  
48 al., 1997). Yet, Fe is a key micronutrient for the growth and metabolism of all living organisms  
49 and especially phytoplankton for which it is essential for the proper functioning of the  
50 photosynthetic system (Behrenfeld & Milligan, 2013; Raven et al., 1999). Consequently, Fe has  
51 a direct influence on primary production (Martin et al., 1994; Sunda & Huntsman, 1995) and  
52 thus plays an important role on carbon export and sequestration in the ocean interior (Martin,  
53 1990). Numerous natural fertilization studies have investigated the importance of iron, primarily  
54 in the Southern Ocean (Blain et al., 2007; Pollard et al., 2007), and have reported enhanced  
55 primary production rates and particulate organic carbon (POC) export efficiencies, which may

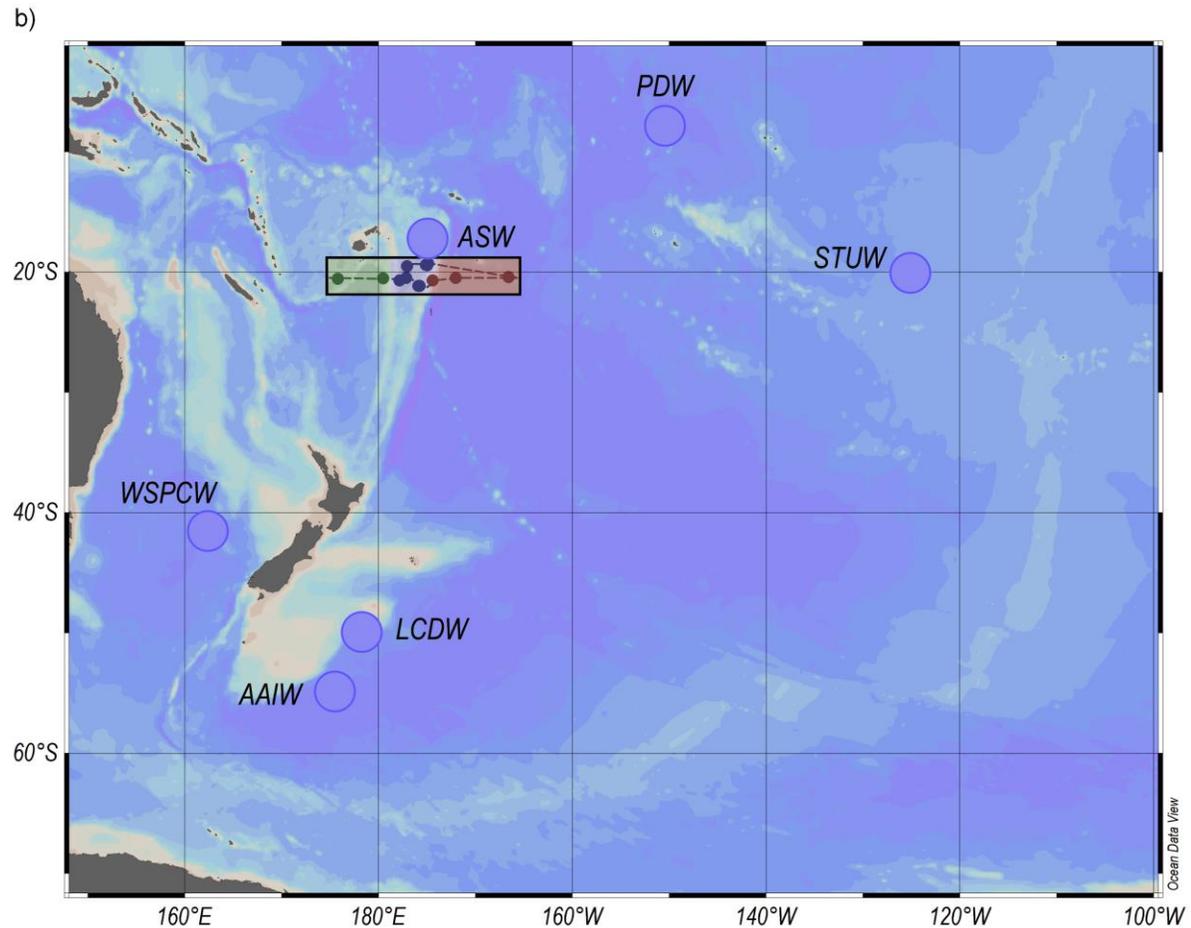
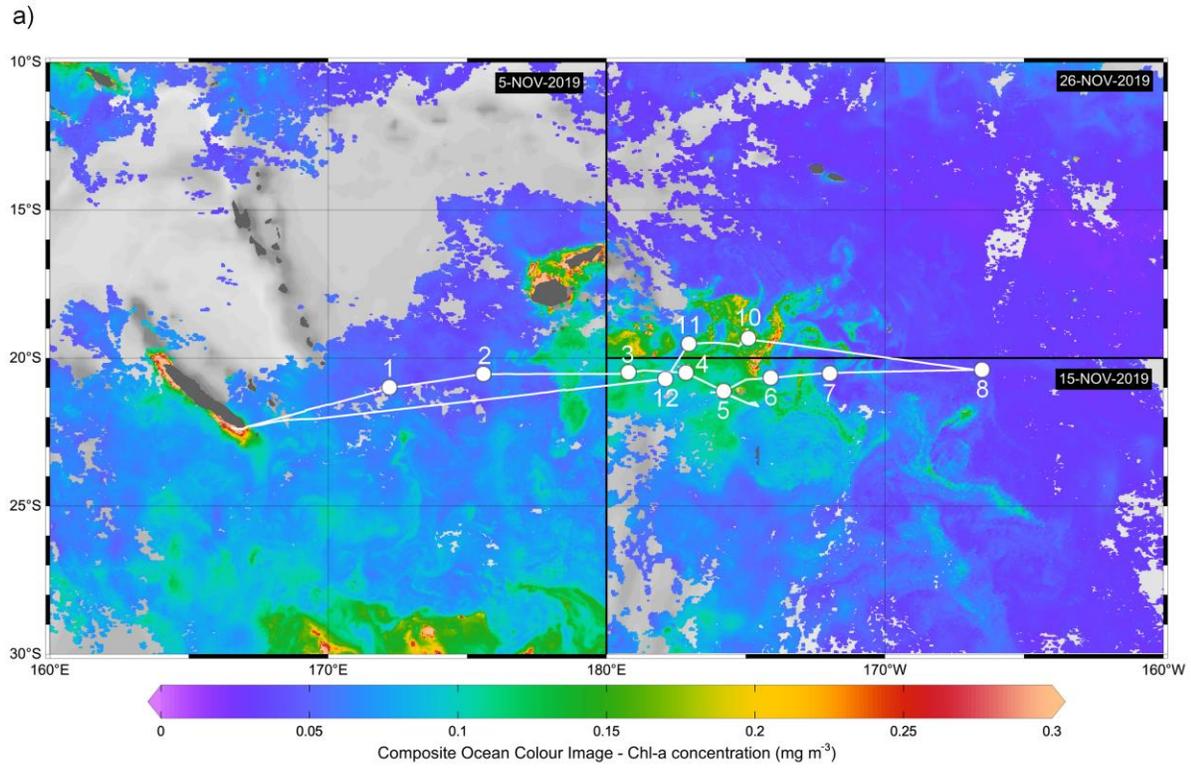
56 influence the biological carbon pump (Morris & Charette, 2013). In the context of climate  
57 change (IPCC, 2021), characterizing the elements governing the efficiency of this pump is of  
58 great interest. Due to its importance, the number of dissolved Fe (dFe) concentration  
59 measurements has increased impressively in recent years thanks to the GEOTRACES program  
60 (<https://www.geotraces.org/>), particularly in the deep ocean. However, there is still a lack of data  
61 for some key ocean regions, such as the Western Tropical South Pacific (WTSP) Ocean.

62 The WTSP Ocean (160 °E to 160 °W) has recently been identified as a hotspot of dinitrogen (N<sub>2</sub>)  
63 fixation with some of the highest rates recorded in the global ocean (Bonnet et al., 2017).  
64 Diazotrophy is a process favored in phosphorus-rich, nitrogen-poor waters and fuels the ocean  
65 with novel nitrogen, helping to maintain ocean productivity and carbon sequestration (Caffin et  
66 al., 2018). This region is characterized by two biogeochemical subregions separated by the  
67 Tonga-Kermadec arc: (1) the South Pacific gyre located east of the arc and characterized by low  
68 N<sub>2</sub> fixation rates (~85 μmol N m<sup>-2</sup> d<sup>-1</sup>) despite sufficient phosphate availability (0.11 μmol L<sup>-1</sup>;  
69 Moutin et al., 2008), and (2) the Melanesian archipelago together with the Lau Basin located  
70 west of the arc and characterized by high N<sub>2</sub> fixation rates (~631 μmol N m<sup>-2</sup> d<sup>-1</sup>; Bonnet et al.,  
71 2018). This spatial decoupling of N<sub>2</sub> fixation has been attributed to the alleviation of dFe  
72 limitation in the Melanesian archipelago (up to 60 nmol L<sup>-1</sup> in the photic layer; Guieu et al.,  
73 2018) in contrast to the dFe-poor waters of the South Pacific gyre (Bonnet et al., 2008; Mills et  
74 al., 2004), with the Tonga-Kermadec arc acting as a frontier separating these two biogeochemical  
75 provinces.

76 The question of the iron origin in the WTSP Ocean remains however open. Multiple external  
77 sources of iron may be good candidates to explain iron availability in Melanesian waters  
78 (Tagliabue et al., 2017). These sources include potential sediment-derived iron inputs in the  
79 shallower waters of the arc, especially near the islands (Dutheil et al., 2018; Homoky et al.,  
80 2016; Raapoto et al., 2019). Similarly, the WTSP Ocean is surrounded by numerous  
81 ferromagnesian islands that may supply many terrigenous nutrients, including iron, through  
82 runoff from island slopes (Shiozaki et al., 2014). Other good candidates are atmospheric iron  
83 inputs emitted from the many aerial volcanoes hosted by the WTSP Ocean. However, these were  
84 quantified during the OUTPACE cruise (Moutin & Bonnet, 2015) and Guieu et al. (2018) stated  
85 that they were too low to explain the dFe concentrations measured in the region. Another  
86 possibility is that dFe may be supplied by water masses of remote origin entering this region

87 (Mahadevan, 2016), therefore the role of physical transport must be also considered. Finally, the  
88 hypothesis that dFe comes from hydrothermal sources was proposed by Guieu et al. (2018) as  
89 they are known to be major sources of iron (German et al., 2016). Such sources are documented  
90 at shallow depths in the region (Massoth et al., 2007) and can directly fuel the photic layer (0-  
91 150 m) with novel dFe. The latter two hypotheses appear to be the most compelling and will be  
92 explored in detail in this work.

93 To understand the sources and sinks affecting the dFe distribution in the WTSP Ocean, we first  
94 present the dFe concentrations measured along a 6100 km transect near  $\sim 20^\circ\text{S}$  (Guieu & Bonnet,  
95 2019; Fig. 1a). We then introduce the theoretical distribution of dFe using a water mass analysis.  
96 The anomalies between theoretical and measured dFe concentrations are then discussed to  
97 highlight the potential hydrothermal origin of dFe in the Lau Basin. Finally, the different  
98 processes involved are discussed to explain the longitudinal evolution of dFe.



100 **Figure 1. (a)** Transect of the cruise superimposed on surface chlorophyll-*a* concentrations ( $\text{mg m}^{-3}$ ). Different  
101 oceanic regions were occupied during the cruise: Melanesian waters including short duration (SD) stations 1, 2 and  
102 3, Lau Basin including SD 4, 11 and 12 as well as long duration (LD) stations 5 and 10, and the South Pacific gyre  
103 including SD 6, 7 and 8. Within each subregion are represented the chlorophyll-*a* concentrations observed over three  
104 time periods corresponding to the mean date of occupancy: 5 November 2019 for the western part of the transect, 15  
105 November for the southeastern part and 29 November for the northeastern part. **(b)** Map of the South Pacific Ocean  
106 showing the transect of the cruise (small dots) and the location at which the end-members were defined (large blue  
107 dots). The frames around the transect represent the different subregions studied during the cruise: green for the  
108 Melanesian waters, blue for the Lau Basin and red for the South Pacific Gyre. See *Table 1* for end-members  
109 acronyms.

## 110 **2. Materials and Methods**

### 111 2.1. Oceanographic campaign presentation and sampling

112 The present study was conducted as part of the TONGA cruise (GPpr14; Guieu & Bonnet, 2019)  
113 that took place onboard the R/V L'Atalante from 31 October to 5 December 2019, along a  
114 transect extending from New Caledonia to the western end of the South Pacific gyre. This 6100  
115 km-long transect allowed the characterization of the two biogeochemical zones of the region  
116 separated by the Tonga-Kermadec arc (Fig. 1). The objective of the cruise was to investigate the  
117 impact of shallow hydrothermal inputs on biogeochemical cycles, particularly trace metal cycles,  
118 downward export, and planktonic communities.

119 *Investigated stations.* Two types of stations were sampled: (a) nine short-duration stations (SD 1,  
120 2, 3, 4, 6, 7, 8, 11 and 12) dedicated to biogeochemical sampling through water column vertical  
121 casts, (b) two long-duration stations (LD 5 and 10) dedicated to the impacts of hydrothermal  
122 fluids from two identified shallow sources along a short ~17 km transect including 5 substations,  
123 named from T5 to T1, with T5 being the closest to the hydrothermal source. For these two long-  
124 duration stations, the hydrothermal sources were identified through a precise dedicated  
125 bathymetric survey, using the one previously reported by Massoth et al. (2007), and the detection  
126 of acoustic anomalies on small caldera-like structures using a multibeam echosounder. The two  
127 T5 substations were positioned where the highest acoustic anomalies were recorded and the  
128 calderas of LD 5 and 10 were identified at 200 and 300 m, respectively. The T1 substations were  
129 then positioned as close as possible to T5 along the main current direction when the bottom was  
130 at ~2000 m for the deployment of a drifting mooring line left for five days. Between T1 and T5,

131 additional stations were staged to investigate the longitudinal impact of hydrothermal fluids  
132 released from T5. Following the eruption of New Late'iki (19.18 °S, 174.87 °W) one month prior  
133 to the cruise (a submarine volcano that turned into an island; Plank et al., 2020), an additional  
134 substation “Proxnov” near this site but located further north (15 km from LD 10-T5) was studied  
135 as part of LD 10.

136 *CTD sampling.* Temperature, conductivity (salinity), and dissolved oxygen vertical profiles were  
137 obtained using a rosette-mounted CTD SBE 9 plus sensor. At each station, (1) conventional CTD  
138 casts were conducted to sample inorganic nutrients using a rosette equipped with 24 Niskin  
139 bottles (12 L) and (2) Trace-Metal clean Rosette (TMR) casts were performed for dissolved and  
140 particulate trace metal sampling (see section 2.2). A total of 398 samples were collected for dFe  
141 analyses. Inorganic nutrients (nitrate:  $\text{NO}_3^-$ , phosphate:  $\text{PO}_4^{3-}$  and silicate:  $\text{Si}(\text{OH})_4$ ) were  
142 measured as detailed in Bonnet et al. (2018).

143 *Autonomous platforms.* Several autonomous instruments were deployed during the cruise at  
144 stations LD 5-T1 and T5, SD 11 and SD 12. Drifter observations were thus possible at the  
145 surface (0-15 m) through the deployment of Surface Velocity Program (SVP) drifters with the  
146 following WMO-id at the stations SD 12 (5501635, 5501636, 5501637, 5501638 and 5501639),  
147 LD 5-T1 (4101779, 4402504, 5102720, 5102721 and 6301680) and LD 5-T5 (5501630,  
148 5501631, 5501632, 5501633 and 5501634; [http://www.coriolis.eu.org/Data-Products/Data-](http://www.coriolis.eu.org/Data-Products/Data-Delivery/Data-selection)  
149 [Delivery/Data-selection](http://www.coriolis.eu.org/Data-Products/Data-Delivery/Data-selection)) and at depth (1000-1500 m) through the deployment of deep Argo  
150 autonomous floats (<https://fleetmonitoring.euro-argo.eu/dashboard>) at the stations LD 5-T1  
151 (6903025), LD 5-T5 (6902985), SD 11 (6902989) and SD 12: 6902927. The short trajectory (5  
152 days, 0-1000 m) of the drifting mooring line deployed at LD 5-T1 could also be observed.

## 153 2.2. Dissolved iron measurements

154 All handling was performed under ultra-clean conditions (Bruland et al., 1979) following the  
155 guidelines established by the GEOTRACES cookbook (Cutter et al., 2017).

156 *Preparation of the sampling material.* All collecting material used was previously washed  
157 following a rigorous procedure. Nalgene® 60 mL low-density polyethylene (LDPE) bottles were  
158 first immersed in a surfactant bath (2% v/v, Decon™) for one week before being rinsed 3-times  
159 with MilliQ water (resistivity > 18.5  $\text{M}\Omega \text{ cm}^{-1}$ ). Bottles were then immersed in a Suprapure

160 hydrochloric acid solution (HCl, Supelco<sup>®</sup>; 10% v/v) for one month before being rinsed again 3-  
161 times with MilliQ water. Vials were finally one-third filled with 1% v/v Ultrapure HCl  
162 (Supelco<sup>®</sup>) and stored double-bagged pending use.

163 *Sampling and storage of seawater samples.* During the cruise, seawater samples were collected  
164 using GO-FLO bottles mounted on a TMR (General Oceanics Inc., Model 1018 Intelligent  
165 Rosette) attached to a 6 mm Kevlar line. Immediately after recovery, the rosette was transferred  
166 into a clean container for sampling. Bottles were pressurized with 0.2 µm-filtered nitrogen (Air  
167 Liquide<sup>®</sup>) allowing in-line filtration on 0.45 µm using acid-cleaned polyethersulfone filter  
168 (Supor<sup>®</sup>). Acid-cleaned bottles were rinsed 3-times with the sampled seawater before the final  
169 collection. Collected samples were acidified to pH 1.7 with Ultrapure HCl (0.2% v/v, Supelco<sup>®</sup>)  
170 within 24 h of collection and stored double-bagged pending analysis.

171 *Dissolved iron analysis in seawater.* Samples for dFe were analyzed in a clean room by flow  
172 injection and chemiluminescence detection (FIA-CL) at the Laboratoire d'Océanographie de  
173 Villefranche, as described by Blain et al. (2008) and adapted from the protocol originally  
174 described by Obata et al. (1997). Some improvements have been made compared to the last  
175 protocol. First, a new 8-hydroxyquinoline (8-HQ) resin was produced according to the protocol  
176 of Landing et al. (1986) where 8-HQ was immobilized on Fractogel TSK HW75-F (Tosoh  
177 Biosciences). The column was manufactured as described by Bowie et al. (1998) (i.e., the resin  
178 was placed in a rigid Teflon<sup>™</sup> tube and isolated with clean quartz wool). Prior to each analysis  
179 day, two successive purifications of luminol on the 8-HQ resin were performed first on the pure  
180 luminol solution and then on the diluted solution. In addition, storage of the luminol solution in  
181 an opaque package significantly improved its shelf life and dosage reproducibility. Finally, the  
182 occasionally observed loss of sensitivity was resolved thanks to a 15-minute aqua regia flush  
183 performed every two analysis days to remove organics aggregated in the 8-HQ system that could  
184 not be eliminated by HCl alone.

185 Apart from these points, the method remained the same as in Blain et al. (2008). Briefly, the  
186 dosage was performed in a closed circuit, which implied no external manipulation and  
187 minimized contamination risk. All tubes used in the analytical system were made of Teflon<sup>™</sup>  
188 except the Tygon<sup>™</sup> tubes used for the peristaltic pump. Since 8-HQ selectively chelates dFe at  
189 pH 5, the pH of the samples was adjusted by adding Ultrapure ammonia (20-22%, Ultrex II, J.T.

190 Baker©) and a 3-times purified ammonium acetate buffer. Samples were then pre-concentrated  
191 on an 8-HQ chelating resin for 120 s. The chelated dFe was eluted with Suprapure HCl (0.7 M)  
192 and mixed with luminol (0.74 M), ammonia (1 M) and hydrogen peroxide (0.7 M). The  
193 luminescent reaction finally produced was detected by a photomultiplier at the end of the circuit.  
194 Given the wide range of measured dFe concentrations ([dFe]), the calibration curves were  
195 adapted to measure [dFe], up to 2 nM for most samples. The dFe-rich samples were diluted up to  
196 1:20 depending on the concentration in a poor-dFe seawater collected at SD 8 also used as an in-  
197 house standard (dFe =  $0.38 \pm 0.03$  nM, n = 26). The final concentration of those diluted samples  
198 did not exceed 5 nM and a 0-5 nM calibration curve was used in that case.

199 *Validation of the measurements.* Analytical blanks (i.e., reagent and manifold), calibration lines,  
200 and reference material values were determined each analysis day and are reported in Table S1.  
201 The mean analytical blank, calculated from the daily determinations with MilliQ water (n = 19),  
202 was  $21 \pm 22$  pM and the detection limit (i.e., three times the standard deviation on the manifold  
203 blank divided by the calibration slope) was  $16 \pm 7$  pM. Each sample was analyzed in triplicate.  
204 Method accuracy was evaluated daily by analyzing the GEOTRACES Surface (GS) seawater  
205 standard daily after each calibration and every 5-10 samples to validate measurements and  
206 monitor analytical stability. An in-house standard, whose concentration was previously  
207 determined by repeated analyses (n = 26) cross-calibrated with the GS standard, was also  
208 measured with the samples as an additional quality control. GS standard analyses for dFe  
209 averaged  $0.510 \pm 0.046$  nM (n = 24) which compares well with community consensus  
210 concentrations of  $0.546 \pm 0.046$  nM. It has to be noted that several replicates were measured to  
211 ensure the accuracy and reproducibility of the method and are reported in Table S1. All  
212 volumetric concentrations were converted to  $\text{nmol kg}^{-1}$  based on temperature and salinity data  
213 measured at each station and sampling depth.

### 214 2.3. Water mass analysis

215 An optimum multiparameter analysis (OMP) was used to resolve the water mass structure along  
216 the cruise transect (Tomczak, 1999). This method determines an optimal least-squares solution of  
217 a linear model of mixing equations to solve the contribution of selected water masses (i.e., end-  
218 members) as a function of a range of hydrologic parameters. In the equational system, the end-  
219 member contributions are used as variables and the hydrographic properties as parameters. In

220 this work, the available parameters are temperature (T), salinity (S), concentrations of dissolved  
 221 oxygen ( $O_2$ ), nitrate ( $NO_3^-$ ), phosphate ( $PO_4^{3-}$ ) and silicic acid ( $Si(OH)_4$ ). However, the system is  
 222 solved by assuming that all parameters are conservative (i.e., there is no sink or source in the  
 223 ocean interior). This is not acceptable in our case as the selected end-members (see section 2.3.3)  
 224 are distributed throughout the South Pacific where nutrients may be consumed through biological  
 225 uptake and produced through organic matter remineralization and are, by definition, non-  
 226 conservative. To account for these biogeochemical processes, an extended OMP (eOMP) was  
 227 performed using the quasi-conservative parameters NO, PO and SiO for which remineralization  
 228 or biological uptake have no effect (Broecker, 1974):

$$229 \quad PO = [O_2] + R_{O_2/P} * [PO_4^{3-}] \quad (1)$$

$$230 \quad NO = [O_2] + R_{O_2/N} * [NO_3^-] \quad (2)$$

$$231 \quad SiO = [O_2] + R_{O_2/Si} * [Si(OH)_4] \quad (3)$$

232 where  $R_{O_2/P}$ ,  $R_{O_2/N}$  and  $R_{O_2/Si}$  are the Redfield ratios that estimate the number of  $O_2$  moles  
 233 consumed for 1 mole of  $PO_4^{3-}$ ,  $NO_3^-$  and  $Si(OH)_4$  released during the process of organic matter  
 234 remineralization, respectively. We assumed a  $R_{O_2/P} = 155$  (Anderson & Sarmiento, 1994),  
 235  $R_{O_2/N} = 9.68$  (Broecker, 1974; Peters et al., 2018) and  $R_{O_2/Si} = 10.33$  (Redfield et al., 1963). This  
 236 led to the following constraint equations:

$$237 \quad x_1 T_1 + x_2 T_2 + \dots + x_n T_n = T_{sample} + \varepsilon_T \quad (4)$$

$$238 \quad x_1 S_1 + x_2 S_2 + \dots + x_n S_n = S_{sample} + \varepsilon_S \quad (5)$$

$$239 \quad x_1 PO_1 + x_2 PO_2 + \dots + x_n PO_n = PO_{sample} + \varepsilon_{PO} \quad (6)$$

$$240 \quad x_1 NO_1 + x_2 NO_2 + \dots + x_n NO_n = NO_{sample} + \varepsilon_{NO} \quad (7)$$

$$241 \quad x_1 SiO_1 + x_2 SiO_2 + \dots + x_n SiO_n = SiO_{sample} + \varepsilon_{SiO} \quad (8)$$

$$242 \quad x_1 + x_2 + \dots + x_n = 1 + \varepsilon_x \quad (9)$$

$$243 \quad x_i \geq 0 \quad (10)$$

244 where  $x_n$  denotes the contribution of the  $n$ th end-member and  $\varepsilon$  refers to residual values that  
 245 account for both measurement error and uncertainty in the assignment of end-member properties.

## 246 2.3.1. Overdetermined system

247 The eOMP must be realized as an overdetermined system, which means that the number of end-  
248 members must be strictly inferior to the number of parameters. We defined six end-members  
249 contributing to the cruise transect (see section 2.3.3) but only five parameters were available (T,  
250 S, NO, PO and SiO). The transect was therefore divided into two vertical domains in which the  
251 eOMP was performed independently, as it has been done in previous studies (e.g., Artigue et al.,  
252 2020; Fitzsimmons et al., 2016): (1) an upper domain comprising depths from 100 to 1000 m  
253 (density from  $23.8 \text{ kg m}^{-3}$  to  $26.8 \text{ kg m}^{-3}$ ), and (2) a lower domain comprising depths from 1000  
254 m to the seafloor ( $> 26.8 \text{ kg m}^{-3}$ ). The eOMP was performed using the OMP v2.0 MatLab  
255 package developed by Johannes Karstensen and Matthias Tomczak (<https://omp.geomar.de/>).

## 256 2.3.2. Upper layer inclusion

257 To consider depths including the shallow sources (0-200 m) in the analysis although the  
258 parameters could not be considered as conservative due to potential biological uptake, an  
259 artificial water mass called Artifactual Surface Water (ASW) was created using salinity data  
260 obtained during the cruise at ~100 m. The limiting depth included in eOMP was selected when  
261 acceptable residuals were obtained ( $< 5\%$ ), leading to the exclusion of water masses above 100  
262 m ( $< 23.8 \text{ kg m}^{-3}$ ). Since the excluded area is the area of maximum interest for surface  
263 communities, the inferred contributions and anomalies in the layers between 100 and 200 m are  
264 assumed not to be affected by biological processes.

## 265 2.3.3. End-member selection and data acquisition

266 End-member identification was performed based on hydrographic properties (potential density  
267 anomaly, salinity, oxygen and nitrate concentrations) observed along the transect profiles (Fig.  
268 S1) and the boundaries of each end-member zone were carefully defined by a thorough review of  
269 the literature. To present “pure” characteristics, end-members properties (Table 1) were selected  
270 from the core of the water masses from several databases (World Ocean Atlas, GLODAP and  
271 World Ocean Circulation Experiment) but were also found in the literature (Blain et al., 2008;  
272 Loscher et al., 1997; Tagliabue et al., 2012). End-members [dFe] were estimated using the  
273 GEOTRACES database. As the South Pacific is largely undersampled, especially for metals,  
274 only one data was available for the Antarctic Intermediate Water (AAIW). For quality purposes,

275 more data ( $n = 85$ ) were found in the Antarctic Circumpolar Current where AAIW flows before  
 276 reaching Tasmanian waters (Bostock et al., 2013) and resulted in a more accurate value for  
 277 AAIW from  $0.478 \pm 0.000 \text{ nmol kg}^{-1}$  ( $n = 1$ ) to  $0.426 \pm 0.123 \text{ nmol kg}^{-1}$  ( $n = 86$ ).

278 **Table 1.** End-members characteristics (value  $\pm$  uncertainty) used for the eOMP.

279 *See the separate Excel file entitled “Table 1” or the table inserted at the end of the manuscript.*

Acronym	Water mass name	T (°C)	S	[O <sub>2</sub> ] ( $\mu\text{mol kg}^{-3}$ )	[PO <sub>4</sub> ] <sup>3-</sup> ( $\mu\text{mol kg}^{-3}$ )	[NO <sub>3</sub> ] <sup>-</sup> ( $\mu\text{mol kg}^{-3}$ )	[Si(OH) <sub>4</sub> ] ( $\mu\text{mol kg}^{-3}$ )	*PO* ( $\mu\text{mol kg}^{-3}$ )	*NO* ( $\mu\text{mol kg}^{-3}$ )	*SiO* ( $\mu\text{mol kg}^{-3}$ )	[dFe] ( $\text{nmol kg}^{-3}$ )	Data source
ASW	Artificial Surface Water	24.000 $\pm 0.750$ $n=29$	34.450 $\pm 0.020$ $n=29$	210.000 $\pm 8.700$ $n=29$	0.100 $\pm 0.100$ $n=12$	0.000 $\pm 0.610$ $n=12$	1.000 $\pm 0.550$ $n=13$	225.500 $\pm 24.200$	210.000 $\pm 14.604$	220.330 $\pm 14.381$	0.180 $\pm 0.030$ $n=9$	TONGA cruise, all stations, November 2019, 19-21 °S, 175 °E-165 °W, 100 m
STUW	Subtropical Underwater	24.464 $\pm 0.833$ $n=27$	36.299 $\pm 0.136$ $n=21$	208.535 $\pm 5.040$ $n=28$	0.177 $\pm 0.030$ $n=35$	0.117 $\pm 0.078$ $n=21$	1.416 $\pm 0.260$ $n=28$	235.984 $\pm 9.619$	209.669 $\pm 5.797$	223.171 $\pm 7.732$	0.134 $\pm 0.018$ $n=8$	World Ocean Atlas, World Ocean Circulation Experiment, Blain et al. (2008), 18-22 °S, 115-135 °W, 20-170 m
WSPCW	Western South Pacific Central Water	10.612 $\pm 0.994$ $n=241$	34.886 $\pm 0.037$ $n=248$	223.926 $\pm 12.560$ $n=239$	0.915 $\pm 0.172$ $n=233$	9.734 $\pm 4.113$ $n=212$	5.839 $\pm 1.97$ $n=232$	365.757 $\pm 39.215$	318.15 $\pm 52.375$	284.247 $\pm 32.959$	0.440 $\pm 0.123$ $n=19$	GEOTRACES GPY01, World Ocean Atlas, World Ocean Circulation Experiment, 39-43 °S, 155-175 °E, 250-500 m
AAIW	Antarctic Intermediate Water	4.317 $\pm 0.583$ $n=96$	34.258 $\pm 0.037$ $n=100$	232.836 $\pm 17.416$ $n=73$	1.924 $\pm 0.143$ $n=76$	28.533 $\pm 2.104$ $n=65$	24.486 $\pm 6.538$ $n=52$	531.076 $\pm 39.522$	509.034 $\pm 37.78$	485.786 $\pm 84.959$	0.426 $\pm 0.123$ $n=86$	GEOTRACES GPY01-06, World Ocean Atlas, World Ocean Circulation Experiment, 53-57 °S, 170 °E-180 °W, 500-1000 m
PDW	Pacific Deep Water	1.952 $\pm 0.142$ $n=171$	34.662 $\pm 0.011$ $n=170$	127.324 $\pm 11.914$ $n=171$	2.713 $\pm 0.090$ $n=171$	37.488 $\pm 0.977$ $n=171$	140.151 $\pm 6.558$ $n=171$	547.808 $\pm 25.801$	490.206 $\pm 21.371$	1575.087 $\pm 79.662$	0.607 $\pm 0.072$ $n=106$	GEOTRACES GP16, World Ocean Atlas, World Ocean Circulation Experiment, 7-11 °S, 160-140 °W, 2000-3000 m
LCDW	Lower Circumpolar Deep Water	0.914 $\pm 0.061$ $n=119$	34.705 $\pm 0.003$ $n=117$	209.459 $\pm 1.731$ $n=103$	2.133 $\pm 0.062$ $n=113$	32.118 $\pm 0.203$ $n=114$	121.569 $\pm 1.885$ $n=113$	540.061 $\pm 11.291$	520.365 $\pm 3.700$	1465.274 $\pm 14.381$	0.635 $\pm 0.178$ $n=8$	World Ocean Atlas, World Ocean Circulation Experiment, Loscher et al. (1997), Tagliabue et al. (2012), 48-52 °S, 172 °E-168 °W, 4000-5000 m

280

### 281 2.3.4. Parameters weighting

282 Parameters were weighted according to their signal-to-noise ratios (measurement accuracy) and  
 283 conservative nature (conservative or quasi-conservative). The highest weight was set for T and S  
 284 and mass conservation was assigned the same weight as the parameter with the highest weight  
 285 (Tomczak & Large, 1989). In this study, different weights were tested for nutrients, ranging from  
 286 values 12 to 4 times lower than T, S and mass conservation. Thereby, T, S and mass  
 287 conservation were adjusted to 24 while nutrient weights were fixed to 4 as these weights led to  
 288 the lowest residuals (Fig. S2).

### 289 2.4. Lagrangian particle tracking experiment

290 Lagrangian Particle Tracking Experiment (LPTE) was conducted to determine the main origin of  
 291 the water masses crossing the cruise transect and thus to ensure the robustness and reliability of  
 292 the defined end-member zones (Fig. 1b). The Ariane Lagrangian analysis software  
 293 (<http://www.univ-brest.fr/lpo/ariane>) and a numerical dataset from a global ocean circulation  
 294 model were used to perform this analysis. The dataset and LPTE method used in this study are  
 295 described in the supporting information (see Text S1).

## 296 2.5. Statistical analyses

297 For [dFe], differences between stations as well as between different depth ranges were tested  
298 using a non-parametric Wilcoxon signed-rank test (*wsrt*), due to the non-independent and non-  
299 normally distributed samples. This test was performed using the R function "wilcox.test".  
300 Differences were considered significant at  $p < 0.05$ .

301 Iron anomalies along the transect were obtained by subtracting measured [dFe] during the cruise  
302 from theoretical [dFe] deduced from the eOMP. Permutation tests were performed to define a  
303 significance level above which the difference obtained actually represents an anomaly. The test  
304 used was the Monte-Carlo analysis and estimated the extent to which dFe anomalies could be  
305 affected by analytical errors of both FIA-CL and eOMP, as well as by the variability in end-  
306 members [dFe]. Thus, 1000 permutations of end-member dFe values were performed according  
307 to their standard deviation to estimate the propagated errors for each dFe anomaly. Since the  
308 highest error was  $\sim 0.2 \text{ nmol kg}^{-1}$ , the anomaly threshold value was set at this value.

309 **3. Results**

## 310 3.1. Dissolved iron distribution in the WTSP Ocean

311 Sections of the [dFe] distribution are presented in Fig. 2, for the cruise transect (a) and for the  
312 small transects above the two shallow hydrothermal sources (b, c). Individual dFe vertical  
313 profiles are shown in Fig. 3 for SD (a, b, c) and LD (d, e, f) stations.

## 314 3.1.1. South Pacific gyre: east of the Tonga-Kermadec arc

315 The eastern part of the transect includes SD 6, 7, and 8 for which we can observe some  
316 differences in the dFe distribution. Stations furthest from the Tonga-Kermadec arc (i.e., SD 7 and  
317 8) were characterized by low [dFe] in the upper layer ( $< 0.2 \text{ nmol kg}^{-1}$ ). Deeper, below 1000 m, a  
318 [dFe] enrichment was visible ( $0.5\text{-}0.6 \text{ nmol kg}^{-1}$ ) down to 3000 m for SD 7 and down to 4000 m  
319 for SD 8. The station closest to the arc (i.e., SD 6) was slightly different from the other stations  
320 of the gyre. Although [dFe] remain low throughout the whole water column ( $< 0.3\text{-}0.4 \text{ nmol kg}^{-1}$ ), [dFe] were higher than those at SD 7 and 8 (*wsrt*,  $p$ -value: 0.03), particularly in the first 100  
321 m of the water column (*wsrt*,  $p$ -value:  $5.28 \cdot 10^{-6}$ ).



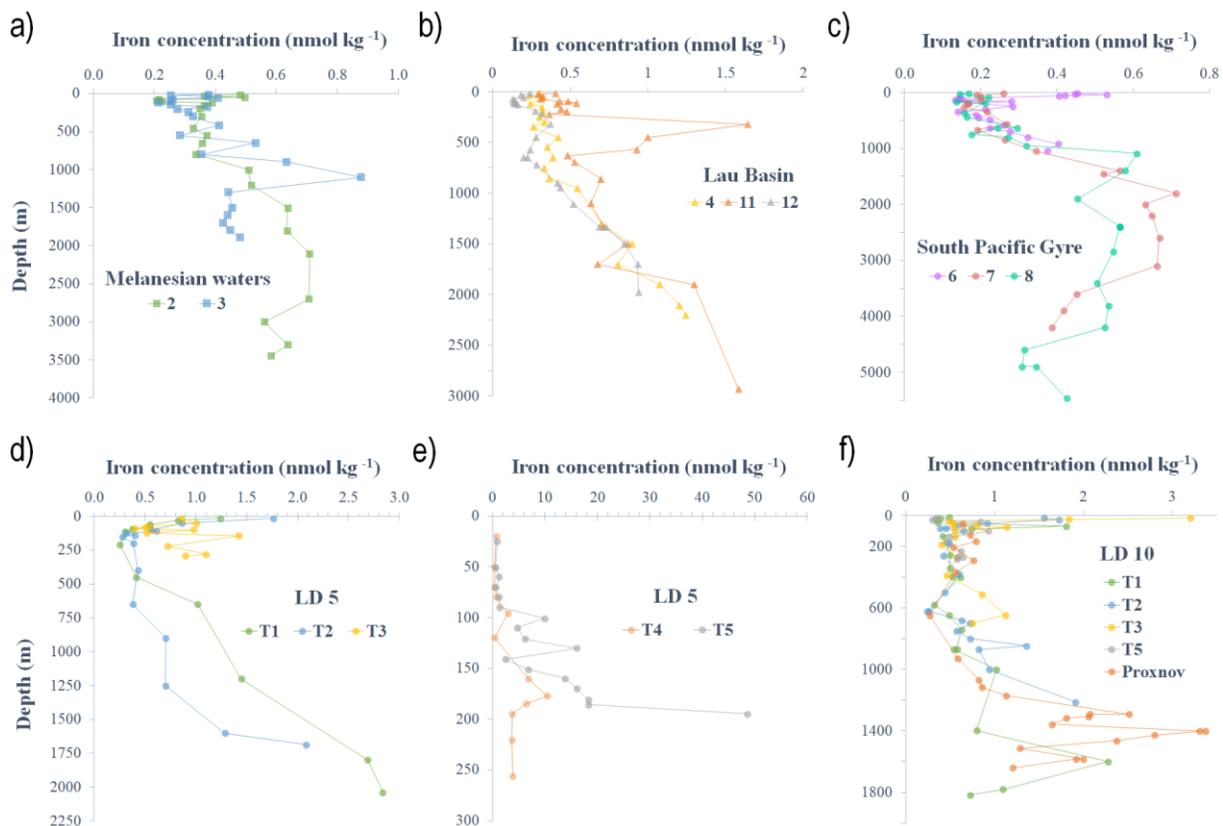
332 **Figure 2.** Sections of the dissolved iron (dFe) concentrations ( $\text{nmol kg}^{-1}$ ) along the cruise transect (a) and along two  
 333 smaller scale transects above LD 5 (b) and LD 10 (c). The dots on the maps represent the position of the sampled  
 334 stations and are colored according to the sub-region in which they are located: green for Melanesian waters, blue for  
 335 the Lau Basin and red for the South Pacific gyre.

### 336 3.1.3. Lau Basin

337 *SD stations.* The Lau Basin, located between the Lau and Tonga ridges, is the central part of the  
 338 transect and includes SD 4, 11, and 12 as well as LD 5 and 10 where two shallow hydrothermal  
 339 sources were studied. In this oceanic basin, we observed higher [dFe] than in the South Pacific  
 340 gyre (*wsrt*, p-value: 0.003) as well as in Melanesian waters (*wsrt*, p-value: 0.04) for all stations  
 341 and over the entire water column. As for the other stations in the transect, some local differences  
 342 were depicted for each profile. The westernmost station of the Lau Basin (i.e., SD 4) was  
 343 characterized by [dFe] of 0.3-0.4  $\text{nmol kg}^{-1}$  between 0 and 800 m. Below 800 m, an enrichment  
 344 is observed with a maximum [dFe] of 1.2  $\text{nmol kg}^{-1}$  at ~2200 m. SD 12 presented the least  
 345 enriched photic layer, with [dFe] similar to those observed to the east (~0.2  $\text{nmol kg}^{-1}$ , *wsrt*, p-  
 346 value: 0.54). At depth, a gradual increase of [dFe] to ~1  $\text{nmol kg}^{-1}$  was observed from 600 m to  
 347 the seafloor. SD 11 had the highest [dFe] of the SD in the Lau Basin. In the photic layer, high  
 348 [dFe] were observed (0.4-0.6  $\text{nmol kg}^{-1}$ ). As for SD 12, a gradual increase of [dFe] up to 1.6  
 349  $\text{nmol kg}^{-1}$  was observed from 600 m to the seafloor. For both stations, a peak centered at ~320 m  
 350 was visible, relatively low for SD 12 (0.4  $\text{nmol kg}^{-1}$ ) and rather high for SD 11 (1.6  $\text{nmol kg}^{-1}$ ).

351 *LD 5.* At LD 5, five substations were investigated: T5, located where the highest acoustic and  
 352 chemical anomalies were found, and T4, T3, T2 and T1, which were located at 0.6, 2, 10 and 17  
 353 km from T5, respectively (Figs. 2b and 3d, e). At T5, high [dFe] (~50  $\text{nmol kg}^{-1}$ ) were observed  
 354 5 m above the seafloor (195 m), decreasing rapidly to ~18  $\text{nmol kg}^{-1}$  at 185 m. These high [dFe]  
 355 were maintained around 10-18  $\text{nmol kg}^{-1}$  up to 100 m before decreasing in the photic layer to a  
 356 [dFe] of ~1  $\text{nmol kg}^{-1}$ . At T4, [dFe] were high in the photic layer, ranging from 0.4 to 2.9  $\text{nmol}$   
 357  $\text{kg}^{-1}$  and even higher at depth (~4-6  $\text{nmol kg}^{-1}$ ). Two [dFe] peaks were observed at 100 and 175  
 358 m. Shear profiles were observable at T3 from surface to bottom, with [dFe] fluctuating between  
 359 0.4 and 1.4  $\text{nmol kg}^{-1}$  and a high [dFe] was visible just at the surface (0.8-1  $\text{nmol kg}^{-1}$  at 0-50 m).  
 360 The profiles of T1 and T2 were quite similar, characterized by high [dFe] in the first 100 m of  
 361 the water column (0.5-2  $\text{nmol kg}^{-1}$ ). [dFe] remained constant at ~0.5  $\text{nmol kg}^{-1}$  between 200 and  
 362 600 m before gradually increasing to the seafloor (2-2.5  $\text{nmol kg}^{-1}$ ).

363 LD 10. As for LD 5, four substations were surveyed at LD 10: T5, which was located on the edge  
 364 of a small caldera summit, and T3, T2 and T1, which were 2, 8 and 15 km from T5. At T5, high  
 365 [dFe] were observed in the entire profile (average  $\sim 0.6 \text{ nmol kg}^{-1}$ ). However, dFe enrichment  
 366 was not as high as the one observed at LD 5-T5 (*wsrt*, *p*-value: 0.01). At T3, surface [dFe] were  
 367 higher than just above the caldera summit, ranging from 2-3  $\text{nmol kg}^{-1}$  between 0-35 m. Below  
 368 35 m, the profile remained constant at  $\sim 0.5 \text{ nmol kg}^{-1}$ . At T2, [dFe] were still high between 0 and  
 369 80 m (0.5-1.5  $\text{nmol kg}^{-1}$ ). Below 80 m and down to 200 m, [dFe] decreased from 0.5 to 0.2  $\text{nmol}$   
 370  $\text{kg}^{-1}$ , followed by an increase to  $\sim 2 \text{ nmol kg}^{-1}$  from 200 m to the seafloor. At T1, [dFe] were  
 371 stable along the entire profile ( $\sim 0.5 \text{ nmol kg}^{-1}$ ) except for two  $\sim 2 \text{ nmol kg}^{-1}$  peaks observed at  
 372 100 and 1000 m. At Proxnov, [dFe] of  $\sim 0.7 \text{ nmol kg}^{-1}$  were observable throughout the water  
 373 column with maxima below 1000 m (2.5, 3.5 and 2  $\text{nmol kg}^{-1}$  at 1250, 1400 and 1600 m,  
 374 respectively).



375

376 **Figure 3.** [dFe] profiles in Melanesian waters (a), Lau Basin (b), South Pacific gyre (c), in the different substations  
 377 T5 to T1 of LD 5 (d, e) and LD 10 along with Proxnov (f). [dFe] distribution at LD 5-T5 (i.e., the cast above the  
 378 hydrothermal source) and LD 5-T4 are plotted separately (e) due to the high [dFe]. Note the different axis scales.

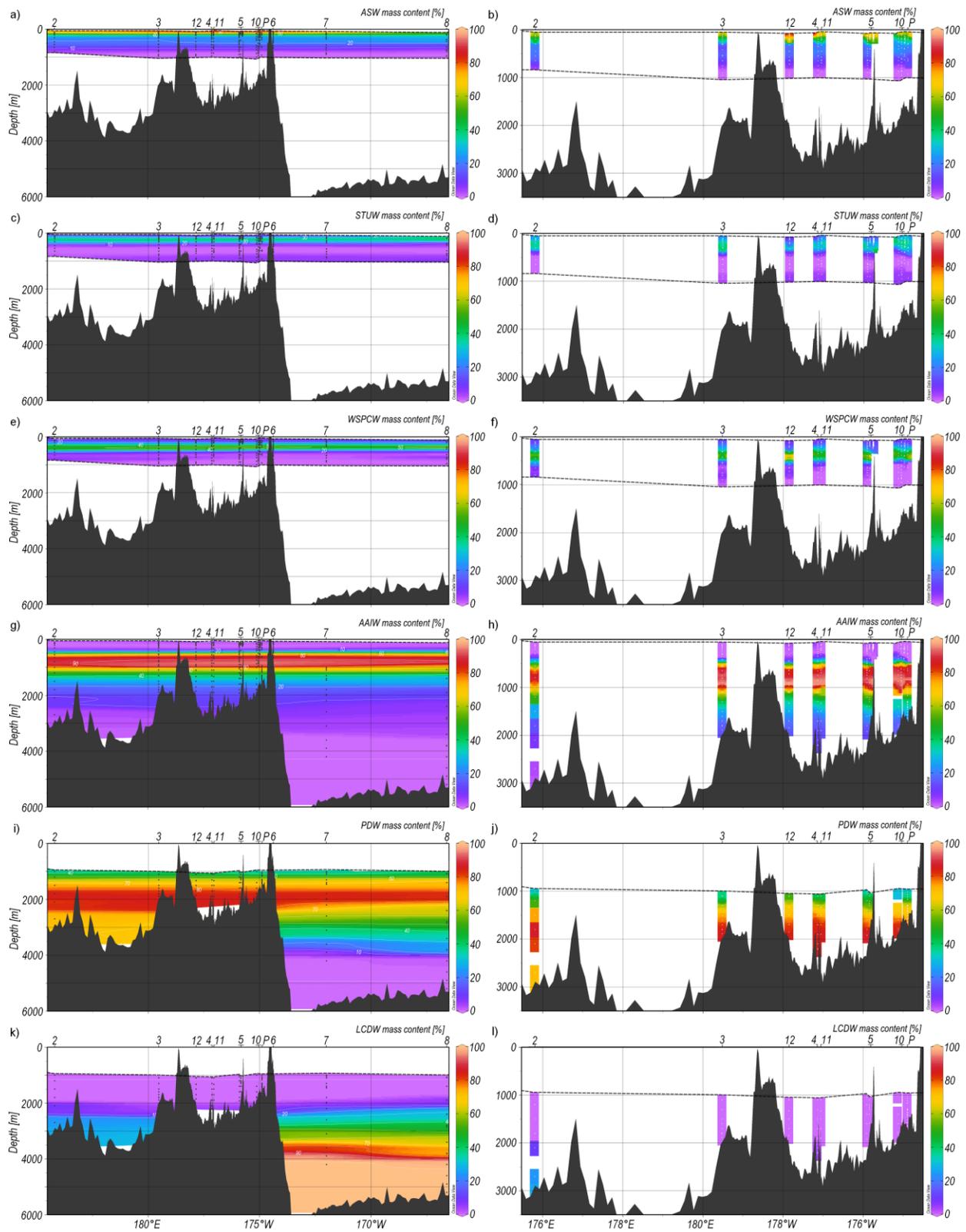
## 379 3.2. Water mass structure in the WTSP Ocean

## 380 3.2.1. Water mass definition

381 The surface layer (100 to 200 m) is characterized by constantly seasonally varying water  
382 properties due to ocean-atmosphere exchanges that have a significant impact on the temperature  
383 and salinity of the water mass (e.g., seasonal temperature variability, subtropical evaporation,  
384 and equatorial excess precipitation). This analytical issue was circumvented by creating the  
385 ASW, which is the only end-member contributing significantly to this layer. The main  
386 thermocline (200 to 700 m) includes the Subtropical Underwater (STUW) and the Western South  
387 Pacific Central Water (WSPCW). STUW originates from the subduction of high salinity waters  
388 from the equatorial part of the subtropical gyre and is associated with a shallow salinity  
389 maximum. Created by subduction and diapycnal mixing, WSPCW exhibits a linear temperature-  
390 salinity relationship over a wide range down to the intermediate layer. The intermediate layer  
391 (700 to 1300 m) was composed solely of AAIW, a low-salinity water mass originating from the  
392 sea surface at sub-Antarctic latitudes and characterized by a salinity minimum reached at 700 m.  
393 AAIW circulates around the subtropical gyre from the Southeastern Pacific, extending  
394 northwestward as tongues of low-salinity, high-oxygen water, and enters the tropics in the  
395 Western Pacific. The deep layer (> 1300 m) contains the Pacific Deep Water (PDW) and the  
396 Lower Circumpolar Deep Water (LCDW). PDW originates from the equatorial Pacific and flows  
397 southward. It is formed in the Pacific interior from the upwelling of the Antarctic Bottom Water  
398 (AABW). PDW is the oldest water in the global ocean, characterized by low oxygen, high nitrate  
399 contents and well-mixed temperature and salinity properties. LCDW originates from the  
400 Southern Ocean and overlaps the depth and density ranges of PDW. It can however be  
401 distinguished from PDW by a maximum of salinity and oxygen and a nitrate minimum. The  
402 property-property profiles of the dataset used for the eOMP and the properties of the defined  
403 end-members are shown in Fig. S3.

404 *A posteriori*, LPTE analyses were performed to ensure that the area chosen for the position and  
405 depth of each end-member agreed with the particle trajectories modeled in the region. The results  
406 of these analyses are shown in Figs. S4, S5 and S6. These analyses support the contribution of  
407 STUW to the thermocline layer of the cruise transect with particles following the South Pacific  
408 gyre circulation between 200-400 m, reaching the selected area for this water mass. Trajectories

409 are also in good agreement with the zone defined for WSPCW as particles, originating from the  
410 Southern Ocean, cross the Tasman Sea between 200 and 400 m before reaching the area of the  
411 cruise transect. LPTE trajectories were consistent with the current understanding of AAIW  
412 circulation: AAIW enters the Pacific Ocean from Eastern New Zealand between 700 and 1500 m  
413 and flows to the WTSP via several portions of water extending from the subtropical gyre,  
414 originating mainly from the Southeastern Pacific. The particle trajectories were also in  
415 agreement for PDW, as particles from the equatorial Pacific reach the PDW-defined zone at  
416 1500-2500 m and flow towards the transect. Regarding LCDW, the LPTE analysis was  
417 consistent with the end-member area chosen for the eastern part of the transect, as particles from  
418 the Southern Ocean flow east of New Zealand before heading northwest and reaching the area of  
419 the transect. This analysis also corroborates the lack of LCDW contribution to the western part of  
420 the transect as demonstrated by the eOMP (see section 3.2.2) since no particles originating from  
421 this water mass reach the western part of the transect.



422

423 **Figure 4.** Contributions (%) of the different water masses to the cruise transect at each sampling depth according to  
 424 the extended optimum multiparameter analysis (eOMP) for (a) the Artfactual Surface Water (ASW), (c) the

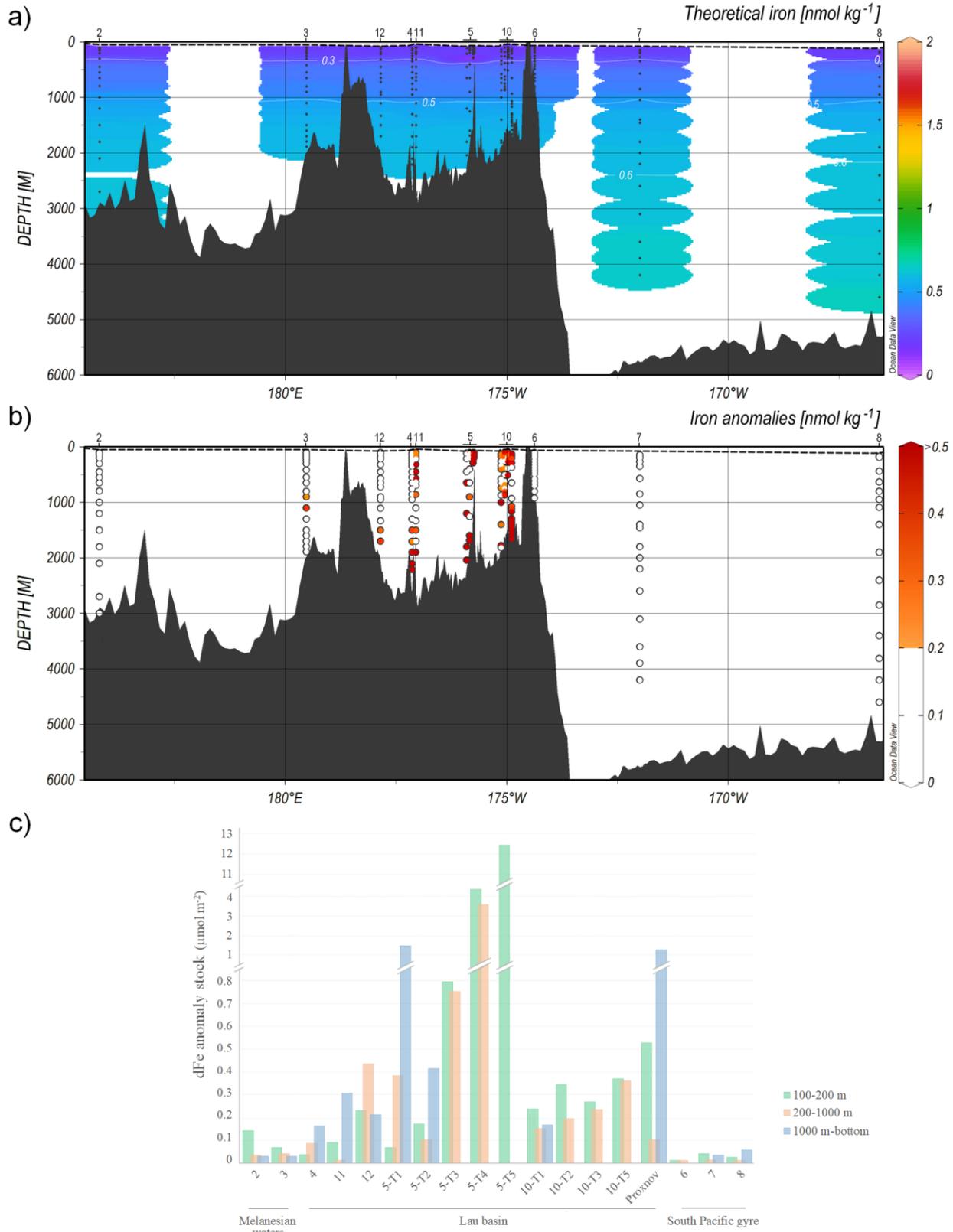
425 Subtropical Underwater (STUW), **(e)** the Western South Pacific Central Water (WSPCW), **(g)** the Antarctic  
426 Intermediate Water (AAIW), **(i)** the Pacific Deep Water (PDW) and **(j)** the Lower Circumpolar Deep Water  
427 (LCDW). The right panels represent the contributions of each water mass in the western part of the transect that  
428 includes Melanesian waters (SD 2 and 3) and Lau Basin (SD 4, 11 and 12; LD 5 and 10) with **(b)** ASW, **(d)** STUW,  
429 **(f)** WSPCW, **(h)** AAIW, **(j)** PDW and **(l)** LCDW. Sampling points are represented by black (left panel) and white  
430 (right panel) dots. The black dotted line represents the boundary above which the eOMP could not be applied (i.e.,  
431 due to non-conservative parameters in the surface layer and residuals > 5%).

### 432 3.2.2. Vertical contribution of water masses along the cruise transect

433 The eOMP results allowed a detailed description of the contribution and distribution of the  
434 selected water masses (i.e., end-members) along the cruise transect (Fig. 4). A uniform zonal  
435 repartition of water masses was visible along the cruise transect, with the exception of the two  
436 deep end-members, PDW and LCDW, for which a different distribution for the eastern and  
437 western parts of the transect was observed. While PDW and LCDW contributed almost equally  
438 to the water masses in the eastern part of the transect below 1000 m, PDW was dominant in the  
439 western part (up to 90%). LCDW thus contributes to only 10-30% of the water masses in the  
440 western part of the transect, which contrasts with its high contribution (up to 100%) in the  
441 deepest waters of the South Pacific gyre.

442 According to the eOMP, the surface layer was mainly occupied by ASW with contributions  
443 greater than 60-90% in the 100-200 m depth range. This contribution decreased rapidly with  
444 depth to less than 30% at ~250 m and reaches zero contribution below 750 m. The thermocline  
445 layer was dominated by two water masses: STUW and WSPCW. STUW was present at depths  
446 between 100 and 450 m and reached its maximum contribution (> 40%) between 150 and 300 m.  
447 Below STUW, a layer of WSPCW was present along the transect with contributions exceeding  
448 60% between 250 and 500 m. The maximum contribution of WSPCW was at ~400 m (60-70%).  
449 AAIW was present over a depth range of 450-1300 m and strongly dominated the intermediate  
450 layer with a contribution exceeding 90% between 700 and 1000 m. Its contribution decreased  
451 rapidly from 80% at 1000 m to 40% at 1300 m. The deep layer was occupied by two water  
452 masses that displayed differences in contribution for the western and eastern parts of the transect.  
453 Mixed with AAIW between 1000 and 1300 m, PDW occupied both intermediate and deep layers.  
454 It dominated the portion between 1300 and 2500 m with a contribution ranging from 60% to  
455 almost 100% in the eastern part of the transect. Its maximum contribution (60-90%) occupied a

456 larger depth range in the western part, as it reached the seafloor of the Melanesian waters (~3000  
457 m) and Lau Basin (~4000 m). In contrast, the deeper seafloor of the South Pacific gyre (~6000  
458 m) was not reached by PDW but by LCDW, a water mass present throughout the eastern section  
459 from 2000 m to the seafloor. Initially mixed with PDW until 3150 m (40%), its contribution  
460 increased to 60-70% between 3200 and 4000 m. LCDW then strongly dominated the deep layer  
461 below 4000 m with a contribution ranging from 90 to 100%. However, in the western part of the  
462 transect, LCDW was only present between 2500 m and the seafloor (~3000 m) in Melanesian  
463 waters and its contribution did not exceed 25%. Its contribution in the Lau Basin was negligible.



465 **Figure 5. (a)** Theoretical dissolved iron (dFe) concentrations along the cruise transect obtained from the extended  
 466 optimum multiparameter analysis (eOMP). **(b)** Iron anomalies representing the difference between measured and  
 467 theoretical dFe concentrations. White dots correspond to iron anomaly values below the  $0.2 \text{ nmol kg}^{-1}$  significance  
 468 threshold. **(c)** Histograms of the dFe anomaly stocks across the region integrated over the surface (100-200 m,  
 469 green), intermediate (200-1000 m, orange) and deep layers ( $> 1000 \text{ m}$ , blue). The order of appearance of the stations  
 470 on the histogram is organized according to the geographic distribution of the sub-regions studied: the Melanesian  
 471 waters (i.e., SD 2 and 3), the Lau Basin (i.e., SD 4, 11 and 12) also including LD 5 and 10 and finally the South  
 472 Pacific gyre (i.e., SD 6, 7 and 8).

### 473 3.3. Iron anomalies along the WTSP Ocean

474 The eOMP-derived dFe transect represents the iron distribution resulting solely from water mass  
 475 transport and physical mixing and acts as a control section without local sources. Those  
 476 theoretical [dFe] were very low and homogeneous across the transect (Fig. 5a) depicting classic  
 477 oceanic iron profiles for each station: low [dFe] in the upper layer ( $0.2\text{-}0.3 \text{ nmol kg}^{-1}$ ) and  
 478 slightly higher [dFe] at depth ( $> 0.4 \text{ nmol kg}^{-1}$ ).

479 Iron anomalies presented in Fig. 5b were obtained by subtracting measured [dFe] (Fig. 2a) from  
 480 theoretical [dFe]. The lowest anomalies ( $\sim 0.09 \text{ nmol kg}^{-1}$ ) below the threshold value were  
 481 observed for SD 6, 7 and 8. In the Melanesian waters, anomalies were higher but still below the  
 482 threshold value, except for a few significant anomalies of  $\sim 0.3 \text{ nmol kg}^{-1}$  observed at 1000 m for  
 483 SD 3. Significant and large anomalies were found for stations located in the Lau Basin both at  
 484 surface and at depth, except for SD 4 and 12 for which anomalies were only present below 1500  
 485 m.

486 DFe stocks attributed to local sources (“anomaly stocks”) integrated over different layers are  
 487 shown in Fig. 5c. No significant differences in anomaly stocks were observed between the three  
 488 depth layers (100-200 m, 200-1000 m, 1000-bottom) for stations located east of the Tonga-  
 489 Kermadec arc (SD 6, 7 and 8) and in Melanesian waters (SD 2 and 3). In contrast, some  
 490 differences were observed in the Lau Basin depending on the depth layer considered, except for  
 491 the westernmost SD 12. Large anomaly stocks were observable in all three layers at SD 12 (up to  
 492  $0.4 \mu\text{mol m}^{-2}$ ) but only in the deep layer for SD 4 ( $0.3 \mu\text{mol m}^{-2}$ ). At LD 5, high anomaly stocks  
 493 were visible in the photic layer from T5 to T3 (from  $12.4$  to  $0.8 \mu\text{mol m}^{-2}$ ) but decreased at T2  
 494 ( $0.17 \mu\text{mol m}^{-2}$ ). In contrast, anomaly stocks in the deep and intermediate layers remained high  
 495 for all LD 5 substations ( $0.4\text{-}3.6 \mu\text{mol m}^{-2}$ ). At LD 10, large anomaly stocks were visible in the

496 surface layer at all substations although they were 10 times lower than at LD 5. At Proxnov, a  
497 large anomaly stock was visible in the photic and deep layers (0.5 and 1.1  $\mu\text{mol m}^{-2}$ ,  
498 respectively).

#### 499 **4. Discussion**

##### 500 4.1. Physical features of the study area

501 Physical processes at play in the region may provide insight into the distribution and fate of  
502 present observations of dFe. The large-scale circulation of the WTSP Ocean is well characterized  
503 (Reid, 1997) and is dominated by the anticyclonic South Pacific gyre. The South Equatorial  
504 Current (SEC) flows westward through the equatorial band and splits into several branches when  
505 it reaches the Lau Basin due to blocking of the SEC by islands associated with the Tonga and  
506 Lau arcs (Webb, 2000). In the western boundary, the SEC splits and joins the East Australian  
507 Current (EAC) that runs along the northwest coast of Australia before flowing eastward to feed  
508 the southern branch of the gyre. It then joins the Humboldt Current that flows northward along  
509 the continental coast of South America (Tomczak & Godfrey, 2003). Gyre waters then return to  
510 the equator through the SEC (Ganachaud et al., 2014). Thus, large-scale circulation patterns of  
511 the study area illustrate the presence of predominant westward currents associated with the SEC.

512  
513 The eOMP coupled with a LPTE performed for this study allowed assessing the structure and  
514 mixing of water masses in the WTSP Ocean and estimates the theoretical distribution of dFe at  
515 the scale of our transect. As most geographic variation in water properties occurs in the  
516 meridional direction in the Pacific Ocean (Talley et al., 2007), it is not surprising that results  
517 showed a quasi-uniform zonal distribution of water masses in the transect conducted sufficiently  
518 far from the EAC along 20 °S. Due to bathymetry sills, deep water masses are not present in the  
519 Lau Basin, as its bowl-shaped seafloor prevents their entry. Thus, the westward-flowing  
520 subtropical gyre appears to collide with the bathymetric barrier represented by the Tonga-  
521 Kermadec arc.

522  
523 Consequences of this bathymetric fracture could be discussed through observations of the surface  
524 and deep trajectories of autonomous instruments (SVP drifters and Argo floats, respectively)  
525 deployed during the cruise both in the center of the Lau Basin and along the Tonga-Kermadec

526 arc. In the center of the Lau Basin (> 100 km downstream of the arc), these trajectories showed  
527 the presence of a southwest flow regardless of the depth (Figs. S7a, c and S8a, c). Numerical  
528 simulations agree with these observations as Rousselet et al. (2018) showed a general transport  
529 west of 170 °W as follows: surface waters enter from the northeast via the SEC and propagate  
530 southwestward towards the various archipelagos present in the WTSP Ocean. Lagrangian  
531 velocities measured along the trajectories are about five times higher at the surface (SVP drifters:  
532 0-15 m) than at depth (Argo floats: 1000-1500 m) in the Lau Basin. Now considering the Tonga-  
533 Kermadec arc (LD 5), Argo float trajectories (Figs. S8b, d) showed a southwestward flow at  
534 depth (1000 m). This observation is also supported by the southwestward trajectory of the  
535 drifting mooring (0-1000 m) deployed at the same site for five days (Fig. 6b). By contrast, at the  
536 surface, a large dispersion of the SVP drifters was observed. Their trajectories showed a  
537 turbulent surface flow (Figs. S7b, d) and the absence of a main stream, in agreement with  
538 Rousselet et al. (2018).

#### 539 4.2. Dissolved iron distribution in the WTSP Ocean

540 This study has provided new insights into the oceanic iron cycle for several reasons. The Pacific  
541 Ocean is severely undersampled, especially for trace metals such as iron (Bruland & Lohan,  
542 2006) and in the WTSP region for which very few data are available (Campbell et al., 2005;  
543 Cohen et al., 2021; Fitzsimmons et al., 2014; Guieu et al., 2018; Massoth et al., 2007). It yet  
544 represents a key region for understanding global dFe dynamics, particularly related to diazotroph  
545 activity. Furthermore, deep hydrothermal iron inputs are widely studied in all ocean regions in  
546 contrast to those induced by shallow hydrothermal vents associated with island arcs (Hawkes et  
547 al., 2014). The Tonga-Kermadec arc hosts a high density of submarine volcanoes associated with  
548 shallow hydrothermal sites (~2.6 vents/100 km; German et al., 2016; Massoth et al., 2007) and  
549 only Guieu et al. (2018) measured elevated [dFe] up to 60 nM in the photic layer and attributed  
550 them to shallow hydrothermal vents. For the first time, a dedicated survey of hydrothermal iron  
551 inputs has been conducted on shallow vents.

##### 552 4.2.1 Iron sources: origin of the dissolved iron inputs

553 Our results demonstrated a low impact of physical dynamics and distant sources on dFe  
554 distribution along the transect. DFe distribution inferred from the eOMP shows low theoretical

555 [dFe] present throughout the transect, including in the Lau Basin. These theoretical [dFe], similar  
556 to open ocean iron data (Blain et al., 2008; Johnson et al., 1997), allow us to conclude that dFe  
557 originates, more probably, from local sources present in the WTSP region.

558 In the eastern part of the transect, the absence of dFe anomalies demonstrates that no local  
559 sources provide iron in the waters of the South Pacific gyre. However, it should be noted that a  
560 dFe enrichment of  $\sim 0.57 \text{ nmol kg}^{-1}$  was measured between 1000 and 3000 m in the gyre (Fig. 2a;  
561 SD 7 and 8). It was initially considered as a deep distal plume from the East Pacific Rise (EPR),  
562 as Resing et al. (2015) measured a plume carrying rich [dFe] ( $\sim 0.75 \text{ nmol kg}^{-1}$  at  $152^\circ \text{W}$ )  
563 thousands of km away from the source (located at  $113^\circ \text{W}$ ). Although this was a more northerly  
564 source, the EPR extends from North America to the tip of South America and hosts numerous  
565 vents that could have a similar impact on the entire ocean basin (Hudson et al., 1986; Menard,  
566 1960). However, this hypothesis has been refuted by the eOMP, which demonstrates the absence  
567 of dFe anomalies to the east, although hydrothermal  $^3\text{He}$  enrichment from the EPR ( $\sim 30\%$ ) was  
568 measured at  $\sim 2500 \text{ m}$  at  $10\text{-}12^\circ \text{S}$ ,  $160\text{-}170^\circ \text{W}$  by Lupton et al. (2004). This suggests that the  
569 anomalies measured by Resing et al. (2015) ultimately merge with the [dFe] of the native deep  
570 water mass ( $0.61 \pm 0.07 \text{ nmol kg}^{-1}$ ) and are no longer visible on this larger scale ( $\sim 6000 \text{ km}$  from  
571 the EPR) with this significance threshold. An absence of anomalies is also observed in  
572 Melanesian waters, except for two anomalies visible at 1000 m at SD 3, suggesting the absence  
573 of local sources in this subregion. It has to be noted that the iron-rich plume centered at  $\sim 180^\circ \text{E}$   
574 as identified by Guieu et al. (2018) at the western border of the Lau arc in March 2015 was not  
575 sampled during our cruise. As hypothesized by the authors, the high [dFe] measured during this  
576 cruise could have been linked to a short-duration mega-plume related to an important submarine  
577 eruption event. The most notable dFe anomalies were found in the Lau Basin, particularly near  
578 the Tonga-Kermadec arc, where hydrothermal sources have already been reported (de Ronde et  
579 al., 2001; Massoth et al., 2007). Strong anomalies are visible both at surface and at depth,  
580 suggesting the presence of one or more local iron sources along this arc system.

581 As described previously, multiple iron sources exist in the open ocean (Tagliabue et al., 2017).  
582 Atmospheric iron inputs can be dismissed, as they were quantified by Guieu et al. (2018) as  
583 being at the lower end of reported values for the remote ocean and could not explain the large  
584 increase observed toward depth. In the WTSP region, four sources can potentially influence the  
585 distribution of dFe. Remineralization can impact the dFe distribution depending on the nature of

586 the particulate iron (pFe) present (Boyd et al., 2010; Bressac et al., 2019). This region is both a  
587 productive zone, rich in biogenic particles favoring remineralization, and a volcanic zone, rich in  
588 lithogenic particles enhancing scavenging. A balance between the two (i.e., source versus sink)  
589 could prevail in the region. However, remineralization has been quantified as redissolving only  
590 1-2% of pFe (Boyd et al., 2010). In addition, Abadie et al. (2017) showed through an iron isotope  
591 study that remineralization occurs preferentially in intermediate waters, with particle desorption  
592 (John & Adkins, 2012; Labatut, 2014) being the primary source of dFe in deep waters. Thus,  
593 remineralization could not explain the dFe anomalies measured both at surface and at depth in  
594 the Lau Basin. Iron inputs from island origin could have a possible impact near the Melanesian  
595 archipelagos. This region hosts many ferromagnesian islands that could supply terrigenous  
596 nutrients, including iron, as hypothesized by Shiozaki et al. (2014), although these fluxes have  
597 not yet been quantified. Similarly, simulations from Dutheil et al. (2018) in the region showed  
598 that sediment-associated iron inputs near island margins could partly control *Trichodesmium*  
599 development. However, the latter two sources would primarily affect the coastal distribution of  
600 dFe at the WTSP Ocean scale and could not explain the observed patterns away from the islands.  
601 Model estimates (Tagliabue et al., 2010) and *in situ* measurements (Fitzsimmons et al., 2014,  
602 2017; Resing et al., 2015) have shown a large-scale impact of deep hydrothermal-derived plumes  
603 on the dFe distribution, up to more than 4000 km off-axis. Such inputs could fertilize the entire  
604 Lau Basin and may explain the dFe anomaly patterns observed in this study. In this context,  
605 other sources seem negligible at the scale of the observed dFe enrichment, especially since it is  
606 remote from the islands.

607 Consistent with this hypothesis, literature and cruise data provide evidence for the hydrothermal  
608 origin of dFe in the WTSP Ocean for several reasons. This region has been reported to host  
609 numerous submarine volcanoes associated with a high density of hydrothermal fields (de Ronde  
610 et al., 2001; Massoth et al., 2007), and of these, two shallow hydrothermal sources were studied  
611 extensively during the cruise. LD 5-T5 appears to be the most active hydrothermal site. It  
612 displays typical chemical and acoustic features, including high turbidity associated with large  
613 amounts of particles, a suboxic environment with low pH (i.e., down to 6.3), low O<sub>2</sub>  
614 concentrations and high levels of H<sub>2</sub>S, CH<sub>4</sub> and CO<sub>2</sub> (Dick et al., 2013). As reported in the  
615 literature in this area (Massoth et al., 2007) and elsewhere (e.g., Dick et al., 2013; González-  
616 Vega et al., 2020; Tarasov, 2006), significant amounts of dFe were released by this hydrothermal

617 site, especially since concentrations up to  $50 \text{ nmol kg}^{-1}$  were measured at 5 m above the seafloor.  
618 This shallow contribution appears to be higher than those from deep sources in the Pacific Ocean  
619 ( $10\text{-}17 \text{ nmol kg}^{-1}$ ; Fitzsimmons et al., 2014; Resing et al., 2015) but in the range of hydrothermal  
620 contributions measured in other oceanic regions (up to  $57 \text{ nmol kg}^{-1}$ ; e.g., Conway & John,  
621 2014). In addition, the highest dFe anomaly stock was recorded at the surface at this site and was  
622 500 times higher than in the South Pacific gyre. Comparatively, LD 10-T5 seems to be a less  
623 active site, with a dFe anomaly stock in the photic layer 35 times lower than LD 5-T5 but still 15  
624 times higher than in the gyre. Multiple clear acoustic anomalies were observed in the multibeam  
625 survey over this hydrothermal site during the cruise, indicating the presence of many weakly  
626 active sources. However, it should be noted that the dFe anomalies recorded at this site do not  
627 appear to be solely of hydrothermal origin, but could also be from the recent submarine eruption  
628 of New Late'iki one month before the cruise (i.e., October 13-23, 2019; Plank et al., 2020). This  
629 assumption seems reasonable since the New Late'iki eruption site, Proxnov and LD 10-T5 were  
630 only 10 and 15 km apart, respectively, and high dFe anomaly stocks were measured in the photic  
631 layer at Proxnov. A large anomaly stock was also estimated in the deep layer for this substation.  
632 Similarly, very abundant volcanic material was collected at 1000 m in sediment traps deployed at  
633 LD 10-T1 (Leblanc, pers. com. 2021) associated with the presence of a deep horizon of turbidity-  
634 identified particles. It can therefore be hypothesized that these large dFe anomalies and high  
635 particle abundance present throughout the water column originate from this submarine eruption.  
636 Thus, the hyperactive volcanism of New Late'iki may have pumped all the energy from the  
637 connected LD 10 system and explain the weakened sources relative to LD 5.

638 The strong dFe anomalies observed near the Tonga-Kermadec arc are thus not limited to the  
639 photic layer. Indeed, anomalies of the same order of magnitude have also been measured in the  
640 deep layer below 1000 m at LD 5-T1 and T2. This suggests the presence of many additional deep  
641 sources along the Tonga-Kermadec arc as reported in the literature (de Ronde et al., 2001;  
642 Massoth et al., 2007), some of them probably located near LD5-T1 where the highest anomaly  
643 stock was measured at depth. Such sources are known to release dFe-rich fluids, sometimes a  
644 million times enriched relative to ambient concentrations in the deep ocean (Von Damm, 1990)  
645 and the emitted plumes can be transported far from the source (Fitzsimmons et al., 2014, 2017;  
646 Resing et al., 2015).

## 4.2.2 Iron sinks: distribution and fate of dissolved iron

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The question of the fate of dFe (i.e., stabilization versus loss) originating from deep and shallow hydrothermalism is also of great importance, particularly due to its influence on the biological carbon pump. Hydrothermal dFe was long thought to be lost near the source, but recent studies have shown that hydrothermal dFe from deep ocean ridges can be transported thousands of kilometers from the source (Fitzsimmons et al., 2014, 2017; Resing et al., 2015; Wu et al., 2011). In agreement with these previous studies, our data show a wide dispersion of deep hydrothermal dFe over several hundred kilometers (~500 km). As mentioned in Section 4.2.1., deep sources appear to be active in the vicinity of the Tonga arc, around 1000 m near LD 5-T1. Thus, anomalies measured further west may be associated with the southwestward dispersion (see Section 4.1) of a deep plume from the arc. This deep iron-rich plume can be traced hundreds of kilometers from the source in the Lau Basin as well as in Melanesian waters, particularly at SD 3 where significant anomalies are visible at ~1000 m. The presence of such high concentrations far from the arc where hydrothermal sources have been identified (de Ronde et al., 2001; Massoth et al., 2007; Stoffers et al., 2006) is due to both advective transport and *in situ* geochemical conditions that allow dFe to remain in solution. The presence of strong iron-binding ligands may explain the stabilization of dFe at depth, especially since ligands have been reported to be abundant in hydrothermal plumes (Bennett et al., 2008; Sander & Koschinsky, 2011). Iron-ligand complexes (Fe-L) are known to enhance the solubility and stabilization of dFe (Boye et al., 2010; Gledhill, 2012; Hering & Morel, 1990; van den Berg, 1995) as they prevent the formation and adsorption of dFe into/onto particles and decrease the reactivity of Fe species (Bennett et al., 2008). Thus, the presence of ligands will mediate the interaction between pFe and dFe in favor of dFe. The formation of unreactive colloidal iron (cFe) could also regulate the stabilization of dFe as it prevents the dFe reactivity with other species or particles (Yücel et al., 2011). It should also be noted that the long residence time of water masses at depth (Fig. S8) should promote the accumulation of dFe near deep vents (Rijkenberg et al., 2018). All these processes could act together to regulate the stabilization of deep hydrothermal iron. However, to our knowledge, the processes governing the stabilization of shallow hydrothermal iron are less well known and should deserve more attention.

676 Compared to deep vents, a weak dispersion of shallow hydrothermal dFe in the WTSP region is  
677 suggested by our data. At LD 5, an almost instantaneous loss of dFe can be observed in the  
678 photic layer: 78% of the initial dFe supply ( $\sim 50 \text{ nmol kg}^{-1}$ ) is lost over a distance of 600 m  
679 between T5 and T4 substations ( $\sim 10 \text{ nmol kg}^{-1}$ ) and almost all dFe (97%) is lost over a distance  
680 of 16 km between T5 and T1 ( $\sim 1 \text{ nmol kg}^{-1}$ ). At LD 10, 47% of the initial dFe input ( $\sim 3.2 \text{ nmol}$   
681  $\text{kg}^{-1}$ ) is lost over a 7 km distance between T3 and T2 substations ( $\sim 1.7 \text{ nmol kg}^{-1}$ ) and 85% of  
682 dFe is lost at T1 ( $\sim 0.5 \text{ nmol kg}^{-1}$ ). Thus, in contrast to deep iron inputs (Bennett et al., 2008;  
683 Fitzsimmons et al., 2014, 2017; Resing et al., 2015), nearly all the dFe supplied by the shallow  
684 hydrothermal source appears to be lost over short distances. On a larger scale, away from the arc,  
685 surface dFe anomalies quickly become similar to those measured in Melanesian waters. Only 0.3  
686 to  $0.5 \text{ nmol kg}^{-1}$  of dFe appears to be stabilized in the photic layer, representing 1 to 9% of the  
687 initial inputs of LD 5 and LD 10, respectively. An exception is observable for the northern SD 11  
688 for which significant dFe anomalies were measured at surface, likely due to the additional impact  
689 from New Late'iki further north.

690 Thus, dFe from shallow hydrothermalism is less spatially spread than dFe from deep  
691 hydrothermalism. The oceanic iron cycle is affected by a series of processes that act together to  
692 set [dFe] in different parts of the ocean (Boyd et al., 2010). Some chemical processes influencing  
693 dFe removal can act in both shallow and deep environments (Johnson et al., 1997; Tagliabue et  
694 al., 2019). It is now widely accepted that most of dFe is rapidly precipitated near hydrothermal  
695 vents due to the rapid mixing of warm,  $\text{H}_2\text{S}$ -rich,  $\text{O}_2$ -poor, low pH fluids with cold,  $\text{O}_2$ -rich, high  
696 pH seawater (Beverkog & Puigdomenech, 1996; Lilley et al., 2013). This causes rapid  
697 precipitation of dFe into iron sulfides or rapid oxidation into iron oxyhydroxide precipitates,  
698 followed by settling on sediments near the source (Bruland & Lohan, 2006). Iron scavenging can  
699 also act in both layers and mediates the removal of dFe by surface adsorption onto sinking  
700 particles or by aggregation of cFe (Balistrieri et al., 1981; Goldberg, 1954; Turekian, 1977). This  
701 process is important in hydrothermal fields as vents release many lithogenic particles that can  
702 enhance the process near the source (Tagliabue & Resing, 2016). However, even though this  
703 process acts throughout the water column, scavenging is likely enhanced in the particle-rich  
704 surface layer as the residence time of dFe relative to scavenging has been reported to be faster in  
705 this layer (10 to 100 days; Black et al., 2020) than at depth (70 to 270 years; Bergquist & Boyle,  
706 2006; Bruland et al., 1979). In addition, biological activities are likely to release high proportion

707 of biogenic particles and simulations from Beghoura et al. (2019) reported a lower sinking rate of  
 708 small inorganic pFe compared to biogenic pFe (up to 2 orders of magnitude). Besides, some  
 709 processes influencing dFe removal act exclusively in shallow environments. For example,  
 710 biological uptake may influence the removal of shallow hydrothermal dFe. Similarly, the  
 711 photochemical reactivity of dFe provides an additional sink relative to deep hydrothermalism.  
 712 The UV portion of the solar spectrum plays a major role in the photoreduction of dFe  
 713 (Rijkenberg et al., 2003), especially when bound to strong ligands. Light-induced reduction leads  
 714 to dissociation of stable Fe-L complexes and unreactive colloids (Johnson et al., 1994; Miller et  
 715 al., 1995). In turn, ligands undergo photo-oxidation and their photodegradation products have a  
 716 lower conditional stability constant (Barbeau et al., 2003), similar to the weak iron-binding  
 717 ligand classes (Rue & Bruland, 1995, 1997). This results in less stable iron species and thus  
 718 ultimately to a dFe loss by scavenging. It should be noted that the short residence time of surface  
 719 water masses (five times faster current velocity, Fig. S7) implies a low accumulation of dFe near  
 720 the shallow source, in contrast to deep plumes.

721 With all these processes in mind and considering a steady state, we developed a simplified 1D 3-  
 722 box budget to quantify the different processes that impact the hydrothermal dFe (Fig. 6a). This  
 723 budget presents the evolution of [dFe] and the processes affecting its distribution vertically and  
 724 horizontally over the small-scale LD 5 transect from T5 to T3. Quantification of each of the  
 725 processes (precipitation, scavenging, photoreduction and biological uptake) was allowed by  
 726 monitoring the evolution of [dFe] over the three substations following these equations:

$$727 \quad P_{min} (\%) = 100 - \left( \frac{[dFe]_{max} * 100}{[dFe]_{input}} \right) \quad (11)$$

$$728 \quad P_{max} (\%) = 100 - \left( \frac{[dFe]_{min} * 100}{[dFe]_{input}} \right) \quad (12)$$

729 where  $P$  represents one of the processes influencing the dFe distribution (precipitation,  
 730 scavenging or photoreduction), with  $P_{min}$  and  $P_{max}$  referring to the minimum and maximum of  
 731 the considered process.  $[dFe]_{input}$  refers to the initial [dFe] of each sub-box.  $[dFe]_{min}$  and  
 732  $[dFe]_{max}$  are the minimum and maximum [dFe] resulting from the processes taking place in  
 733 each sub-box.

734 For dFe biological uptake (BU), we considered the rates measured during the cruise (minimum  
 735 uptake values ( $UR_{min}$ ) = 9; maximum uptake value ( $UR_{max}$ ) = 17 pmol kg<sup>-1</sup> d<sup>-1</sup>; Lory et al.,  
 736 2022). The impact of biological uptake on [dFe] were estimated as follow:

$$737 \quad BU_{min} (\%) = \left( \frac{UR_{min} * 100}{[dFe]_{input}} \right) \quad (13)$$

$$738 \quad BU_{max} (\%) = \left( \frac{UR_{max} * 100}{[dFe]_{min}} \right) \quad (14)$$

739 where  $BU_{min}$  and  $BU_{max}$  are the minimum and maximum [dFe] removed by biological uptake.

740 At LD 5-T5, the water column is divided into three sub-boxes representing different layers in  
 741 which distinct processes come into play: the (1) photic (0-90 m), (2) sub-photoc (90-150 m) and  
 742 (3) suboxic (150-200 m) layers. Sub-box (3) is characterized by high [H<sub>2</sub>S] and decreasing pH  
 743 and [O<sub>2</sub>]. According to our budget, seawater conditions in this layer exclusively promoted the  
 744 rapid precipitation (Lilley et al., 2013) of 86% of dFe into sulfides and/or oxyhydroxides. This  
 745 estimate is in agreement with those reported in the literature for deep vents (32 to 90%; (Field &  
 746 Sherrell, 2000; González-Santana et al., 2020; Lough et al., 2019). A vertical flux of 17 μmol  
 747 dFe m<sup>-2</sup> d<sup>-1</sup> entering the sub-photoc layer was estimated (see Text S2 for methodological details).  
 748 Environmental conditions in the sub-photoc layer no longer favor precipitation, due to stable pH  
 749 and [O<sub>2</sub>] and the absence of H<sub>2</sub>S, and precipitation was considered negligible. In this layer,  
 750 scavenging removed 9-63% of dFe. Since this wide range obtained by monitoring the  
 751 disappearance of dFe was unsatisfying for our budget, this scavenging estimate was finally  
 752 refined for the sub-box (2) by monitoring the appearance of pFe (see Text S3 for methodology of  
 753 pFe measurements) according to the following equations:

$$754 \quad S_{min} (\%) = 100 - \left( \frac{[pFe]_{input} * 100}{[pFe]_{min}} \right) \quad (15)$$

$$755 \quad S_{max} (\%) = 100 - \left( \frac{[pFe]_{input} * 100}{[pFe]_{max}} \right) \quad (16)$$

756 where  $S_{min}$  and  $S_{max}$  refer to the minimum and maximum of scavenging.  $[pFe]_{input}$  being the  
 757 initial [pFe] of each sub-box.  $[pFe]_{min}$  and  $[pFe]_{max}$  are the minimum and maximum [pFe]  
 758 resulting from the scavenging effect.

759 Obtained scavenging percentages were consistent with those estimated with dFe only, although  
760 narrower: 40-72% and 67-68% in sub-boxes (1) and (2), respectively, and of the same order of  
761 magnitude in the two layers. Those scavenging estimates are in good agreement, although  
762 slightly higher, with those reported in the literature (~50%; Forsgren et al., 1996; González-  
763 Santana et al., 2021). Vertical dFe fluxes into the photic layer from the sub-photoc layer were one  
764 order of magnitude lower ( $1.9 \mu\text{mol dFe m}^{-2} \text{d}^{-1}$ ) compared to fluxes from suboxic to sub-photoc  
765 layers but still high enough to allow a significant vertical transport of dFe. In that layer,  
766 photoreduction, biological uptake and scavenging have to be considered. DFe consumption by  
767 surface phytoplankton has been reported to be low in the WTSP region, including above shallow  
768 hydrothermal sources ( $9\text{-}17 \text{ pmol L}^{-1} \text{d}^{-1}$ ; Lory et al., 2022). This uptake rate is similar to that  
769 reported in the literature ( $29 \pm 19 \text{ pmol L}^{-1} \text{d}^{-1}$ ; Maldonado et al., 2005; Sunda & Huntsman,  
770 1995) and accounts for only 0.7-3% of dFe removal. Assuming a similar scavenging rate in the  
771 photic and sub-photoc layers, it was possible to estimate a photoreduction of 22-36% of dFe, in  
772 agreement with values previously reported in the literature ( $25 \pm 21\%$ ; Kuma et al., 1992).

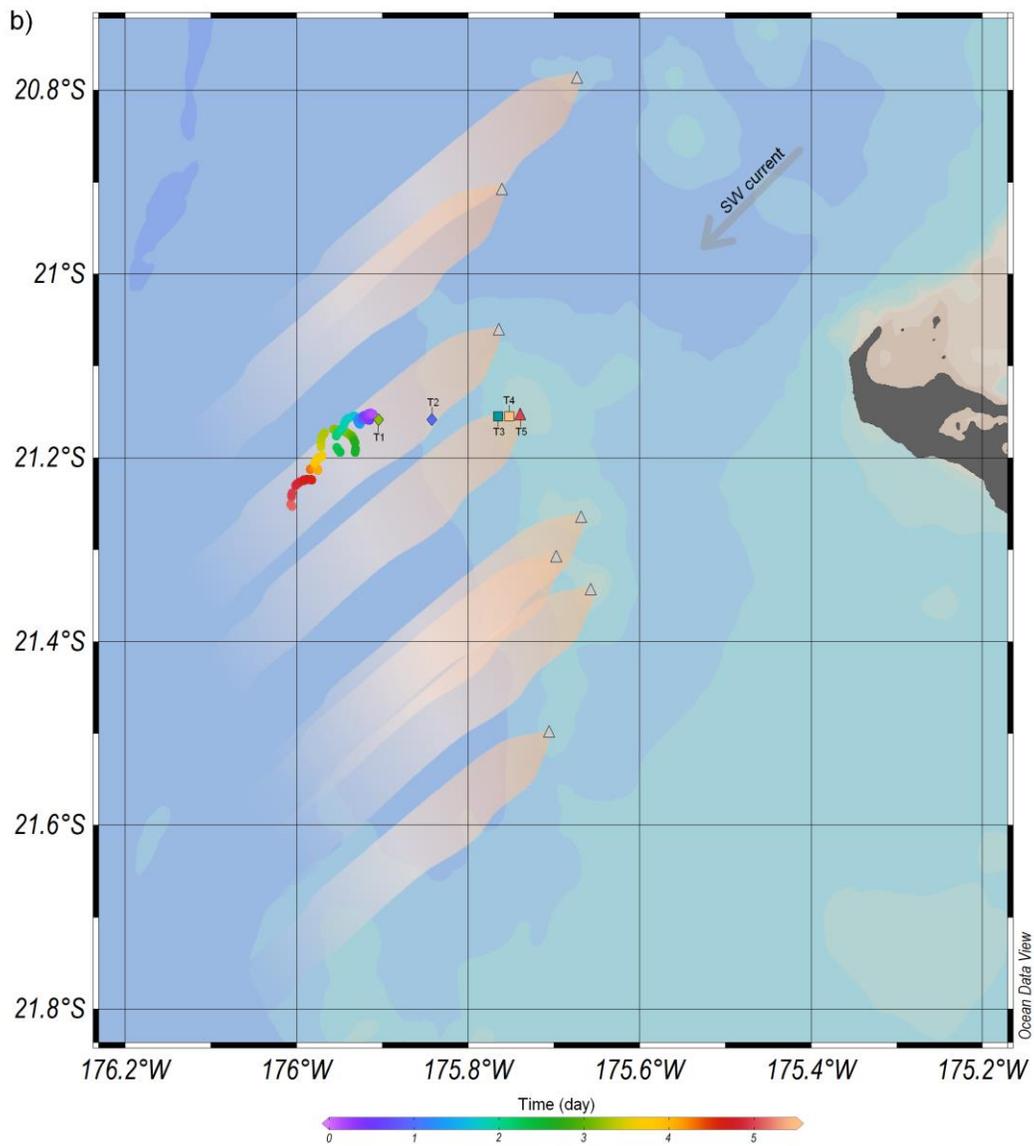
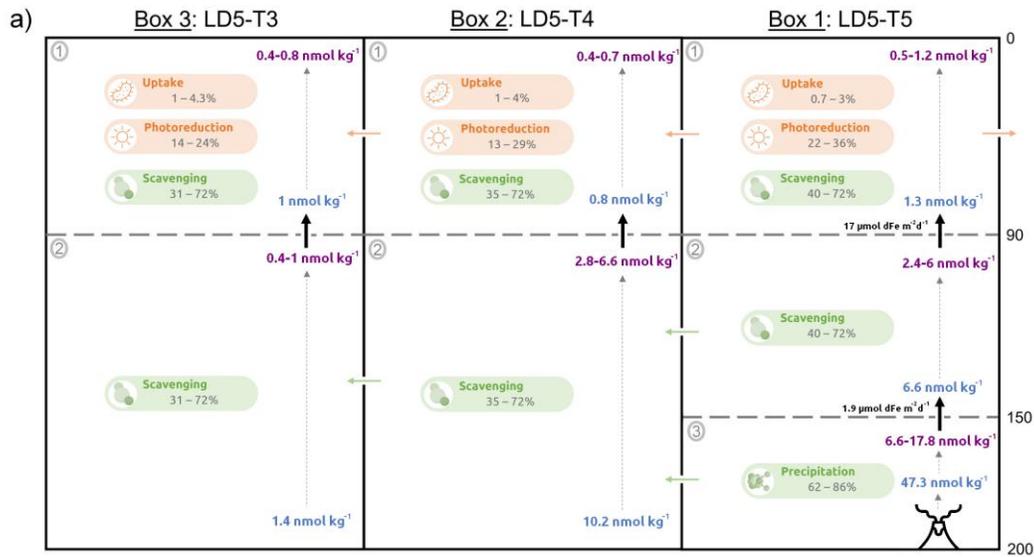
773 The second box refers to the LD 5-T4 substation and has been divided into two sub-boxes to  
774 discriminate processes acting exclusively in the photic layer. Since precipitation is assumed to be  
775 negligible far from the hydrothermal source due to seawater chemical conditions, scavenging is  
776 the main process acting in the sub-photoc layer and was estimated to remove 35-72% of dFe.  
777 Biological uptake and photoreduction processes acting in the photic layer were estimated as  
778 described above, leading to a removal of dFe of 1-4% and 13-29%, respectively. The third box,  
779 divided into two sub-boxes, corresponds to LD 5-T3. Scavenging was estimated to remove 31-  
780 72% in the sub-photoc layer while biological uptake and photoreduction were removing 1-4.3%  
781 and 14-24%, respectively, in the photic layer. It can be noted that the horizontal fluxes of dFe  
782 from T5 towards T4 and T3 could not be quantified due to a lack of accurate characterization of  
783 the vertical flow. Qualitatively, however, the horizontal flux would be diffusive in the photic  
784 layer and below, controlled by southwestward advection (as inferred from Lagrangian  
785 observations, see Section 4.1).

786 This budget indicates that the considered processes appear to act in similar proportion at all  
787 substations. In the photic layer, biological uptake is a negligible process in such iron-rich  
788 environment whereas photoreduction significantly impacts dFe distribution (removing up to 36%  
789 of dFe). In addition, scavenging also plays an important role in decreasing [dFe] in both layers as

790 it removes more dFe from solution (up to 72%) compared with data reported for deep sources  
791 (Forsgren et al., 1996; González-Santana et al., 2020).

792 It can be noted that T2 and T1 substations have been excluded from the budget because no  
793 significant [dFe] anomalies were found at these two substations. An impact of hydrothermal dFe  
794 from T5 was unlikely at these substations as the main stream at depth transported the plume  
795 southwestward (see Fig. 6b and Section 4.1). Nevertheless, the relatively large [dFe] ( $> 0.5 \text{ nmol}$   
796  $\text{kg}^{-1}$ ) observed at these substations could be due to the influence of other active shallow sources  
797 further north of the arc (Massoth et al., 2007).

798 DFe from shallow hydrothermal sources thus appears to be less spatially spread than dFe from  
799 deep sources due to the action of several processes (Johnson et al., 1997; Tagliabue et al., 2019).  
800 An influence of physical processes cannot be denied given the residence time of water masses,  
801 much shorter at the surface than at depth. The accumulation of dFe released by shallow  
802 hydrothermal sources will therefore be much lower than from sources located at depth.  
803 Biological processes do not have a significant impact on the distribution and fate of shallow  
804 hydrothermal dFe since our results showed a negligible effect of this process. Chemical  
805 processes appear to be the most important in mediating the fate of hydrothermal dFe in the photic  
806 layer. Although some of these, such as precipitation and scavenging, act throughout the water  
807 column, scavenging seems to act most effectively in the particle-rich surface layer. In addition,  
808 one mechanism for stabilizing dFe from deep plumes is complexation with ligands. However,  
809 photoreduction occurring in the photic layer implies a dissociation of dFe-stabilizing complexes,  
810 releasing inorganic dFe that can be rapidly removed from the dissolved pool. All these processes,  
811 not occurring in the deep dark ocean or to a lesser extent, act together to remove shallow  
812 hydrothermal dFe on a relatively small spatial scale (i.e., a few kilometers). Nevertheless, the  
813 cumulative impact of other active shallow sources identified in the vicinity of the Tonga-  
814 Kermadec arc (Fig. 6a) should fertilize the entire Lau Basin with dFe.



816 **Figure 6. (a)** Representation of the simplified 1D 3-box budget for substations T5, T4 and T3 of LD 5. The grey  
817 circled numbers in each box correspond to the number of the sub-boxes representing the (1) photic, (2) sub-photic  
818 and (3) suboxic layers. To the right is the depth considered for each sub-box in the budget. [dFe] used for the  
819 budget, namely the dFe input entering in each sub-box (i.e., either by the action of the hydrothermal source (sub-box  
820 (3)) or by physical processes) and the ranges (i.e., min-max) of [dFe] evolving in each sub-box from the initial input  
821 are represented in blue and purple, respectively. Orange cells represent processes acting exclusively in the photic  
822 layer while green cells represent processes that act in all layers. The non-dotted arrows represent transport: black for  
823 vertical diffusion, green for advective transport and orange for horizontal diffusion occurring only in the photic  
824 layer. **(b)** Conceptual diagram representing the dispersion of the measured plume at substations T5, T4 and T3 of  
825 LD 5 and the additional impact of plumes from sources further north or south of our Tonga arc study area on the Lau  
826 Basin. The colored points represent the southwestward trajectory of the drifting mooring (average flow on 0-1000  
827 m) deployed at LD 5-T1 and left drifting for five days.

## 828 **5 Conclusion**

829 The Western Tropical South Pacific Ocean is characterized by the existence of a biogeochemical  
830 boundary between its western part, including the Lau Basin and Melanesian waters and  
831 characterized by high N<sub>2</sub> fixation, and its eastern part, comprising the western end of the South  
832 Pacific gyre and characterized by extremely low diazotrophy rates. Measurements of dFe  
833 concentrations in these two subregions confirm that this spatial decoupling of diazotrophy  
834 patterns can be explained by the attenuation of Fe limitation west of the Tonga-Kermadec arc.  
835 The question of the dFe origin was resolved using the results of an eOMP performed on the  
836 cruise transect. By separating the “conservative” component from the dFe section, it was  
837 possible to estimate the dFe component coming only from local sources, mostly from  
838 hydrothermal sources present in this region. Thus, this study was able to highlight the impact of  
839 shallow hydrothermal sources located along the Tonga-Kermadec arc on the dFe cycle of the  
840 WTSP Ocean. These sources release a large amount of dFe directly into the photic layer and can  
841 fertilize the entire Lau Basin and, to a lesser extent, the Melanesian zone. Such fertilization has a  
842 non-negligible impact on biological communities, particularly on diazotrophs such as  
843 *Trichodesmium* that are known to be widespread in this part of the Pacific Ocean. However,  
844 while dFe-rich plumes from deep sources appear to be stabilized and transported in Melanesian  
845 waters, dFe from shallow sources persists for only a few tens of kilometers. DFe is indeed lost  
846 very rapidly from solution by several physical, chemical, and biological processes such as  
847 precipitation and scavenging, some of them acting exclusively in the photic layer such as

848 photoreduction and, to a lesser extent, biological uptake. This weak stabilization of shallow  
849 hydrothermal dFe results in part from the dissociation of dFe-stabilizing complexes and weak  
850 dFe accumulation mediated by the faster ocean circulation in surface than at depth. Scavenging  
851 has also been reported to be more intense in the surface layer, accelerating the removal of  
852 shallow hydrothermal iron. Thus, dFe from shallow hydrothermal plumes of the Tonga-  
853 Kermadec arc does not appear to be transported over very long distances as previously  
854 demonstrated for deep hydrothermal plumes by many studies. Further analysis of the  
855 geochemical differences that may exist between shallow and deep hydrothermal sources and  
856 their environment is needed to better understand the stabilization of dFe and other metals in this  
857 region. For example, a thorough study of the various ligands released from shallow and deep  
858 hydrothermal sources in the region and an assessment of their stability is needed to understand  
859 the differences between these two types of hydrothermal systems. An investigation of the  
860 behavior of other trace metals released from such sources could help determine whether the low  
861 Fe dispersion observed in shallow hydrothermal environments is a distinct feature of that  
862 element or may be observed for other dissolved metals. In addition, further sampling of the Lau  
863 Basin with finer temporal and/or spatial scales may be necessary for a more thorough study of  
864 the dispersion of iron-rich shallow hydrothermal plumes.

### 865 **Acknowledgments and Data**

866 The entire dataset used in this study is currently available in the LEFE-CYBER database of the  
867 TONGA cruise (<http://www.obs-vlfr.fr/proof/php/TONGA/tonga.php>).

868 This work was performed in the framework of the TONGA project  
869 ([dx.doi.org/10.17600/18000884](https://dx.doi.org/10.17600/18000884)) managed by the LOV (C. Guieu) and the MIO (S. Bonnet) and  
870 funded by the Flotte Océanographique Française, the A-MIDeX of the Aix-Marseille University,  
871 the LEFE-CYBER program and the ANR. We warmly thank all the scientists, the captain and  
872 the crew of the R/V L'Atalante for their cooperative work at sea. We thank H el ene Planquette  
873 for the pFe measurements and for reviewing this study. We thank Sandra Nunige for the  
874 processing of nutrients analyses. We thank Lise Artigue for her ingenious idea to couple eOMP  
875 and biogeochemical analyses.

876 **Competing Interests**

877 The authors declare that they have no conflict of interest.

878 **References**

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